Portable Power: Advanced Rechargeable Lithium Batteries

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Guest Editors

Abstract

The full potential of wireless devices remains unattainable due to limitations in battery performance. It is the thesis of the guest editors and contributing authors of this issue of *MRS Bulletin* that there is much room for improvement, that we are still far from the practical limits of the technology, and that materials research has the capability to pave the way for a new generation of rechargeable batteries that will offer a dramatic improvement in power delivery over anything available today. The basics of battery operation, including the relevant electrochemistry, are reviewed, unsolved problems are enumerated, and prospective solutions are indicated.

Keywords: *electrical properties, energy-storage materials, polymers, rechargeable lithium batteries.*

Introduction

Major advances in materials have paved the way for an array of wireless devices with ever-increasing functionality. Oddly, their full potential remains unattainable, and this is almost exclusively due to the limitations of the batteries that power them. Ironically, it was more than 200 years ago that Alessandro Volta at the University of Pavia demonstrated for the world the first battery, comprising a stack of coin-sized disks of zinc and silver arranged in pairs separated by cardboard soaked in salt water.[1] Yet, battery technology has seemingly been untouched by radical innovation in the decades since then. It is the thesis of the guest editors and contributing authors of this issue of *MRS Bulletin* that there is much room for improvement, that we are still far from the practical limits of the technology, and that materials research has the capability to pave the way for a new generation of rechargeable batteries that will offer a dramatic improvement in power delivery over anything available today.

Rechargeable Batteries: The Basics

There are many chemistries that will serve as the basis for rechargeable, or secondary, batteries.[2] Distinctions can be made on the basis of the following metrics: voltage, current (maximum, steady state, and peak), energy density (watt-hours per kilogram and per liter), power density (watts per kilogram and per liter), service life (cycles to failure), and cost (per kilowatt-hour). Table I summarizes the performance characteristics of some secondary battery types. Such comparisons are made difficult by the fact that many performance characteristics are functions of battery size and service conditions. For example, service life is greatly affected by discharge current and depth of discharge, DOD (a fully charged battery is considered to be at 0% DOD, while a "dead" battery is considered to be at 100% DOD). However, both of these operating parameters can be very different for cells based upon the same chemistry but designed for different applications. With this caveat, Table I is presented.

With its low density (0.53 g cm^{-3}), low electronegativity, and high electron/atom mass ratio, lithium has become the preferred choice for the active element of the anode, which on discharge functions as an electron donor according to

$$\textit{anode:} \qquad x\mathrm{Li} \rightarrow x\mathrm{Li}^{+} + xe^{-} \rightarrow \text{discharge}, \tag{1}$$

where Li^+ enters the electrolyte, and the electron exits the anode to the external circuit to power the load. The elemental lithium is typically present in a host insertion material, most commonly a lithiated carbon such as Li_xC_6.[3] Figure 1 shows a schematic representation of a lithium battery in discharge mode.

Clearly, the electrolyte must be an ionic conductor capable of solvating Li^+ ions. Because lithium is more electropositive than hydrogen, the electrolyte must be nonaqueous and aprotic. A representative formulation is a solution (1:1 by volume) of ethylene carbonate and propylene carbonate containing a suitable lithium salt (at a concentration of about 1 M) such as lithium hexafluorophosphate, $LiPF_6$.[4] For

Table I: Comparison of the Performance Characteristics of Secondary Batteries.

Battery Type	Nominal Voltage (V)	Specific Energy (Wh/kg)	Specific Energy (kJ/kg)	Volumetric Energy (Wh/l)	Volumetric Energy (kJ/l)
Pb-acid	2	35	126	70	252
Ni-Cd	1.2	40	144	100	360
Ni-MH	1.2	90	324	245	882
Ag-Zn	1.5	110	396	220	792
Li-ion	3.6	125	450	440	1584
Li-SPE*	3.1	400	1440	800	2880

Note: From Reference 2.
* Projections based upon thin-film microbattery test results in the laboratory with Li/SPE/VO_x; SPE stands for solid polymer electrolyte.

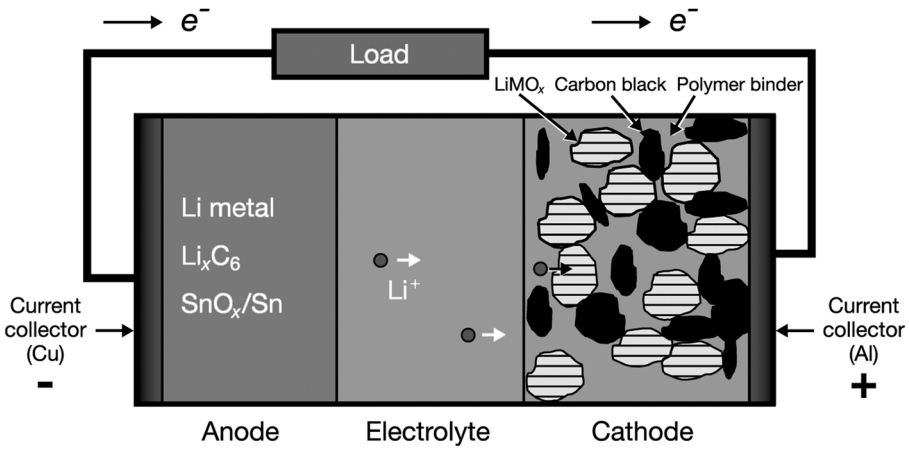

Figure 1. Schematic illustration of a rechargeable lithium battery in discharge mode.

safety, a separator made of a polyolefin such as microporous polypropylene is placed between the electrodes.[5] If the electrolyte temperature exceeds a certain value, the separator melts, closing the pores, and current ceases to flow.

At the cathode, Li^+ engages in an electron-transfer reaction that both decreases the chemical potential of lithium relative to its value in the anode and calls for a compensating electron: on discharge, the cathode functions as an electron acceptor. In commercial cells, the cathode-active material is a lithiated transition-metal oxide[6] such as lithium cobalt oxide, which reacts according to

$$\textit{cathode:} \quad xLi^+ + xe^- + LiCoO_2 \rightarrow Li_{1+x}CoO_2 \rightarrow \text{discharge.} \quad (2)$$

Figure 1 shows $LiMO_2$ mixed with carbon black, which is added to raise the electronic conductivity of the cathode.

Alternatively, the elementary electrochemical reactions can be represented as follows:

$$\textit{anode:} \quad xLi \rightarrow xLi^+ + xe^- \quad (3)$$

and

$$\textit{cathode:} \quad xCo^{4+} + xe^- \rightarrow xCo^{3+}. \quad (4)$$

If the battery is to be rechargeable, the reactions must be reversible; that is, if the load in the external circuit is replaced with a power supply, it should be possible to run the reactions at the cathode and the anode in reverse.

The current–voltage characteristic of a battery is affected by the instant kinetics of operation: the lower the current drain rate, the smaller the difference between theoretical and observed values of voltage and capacity. This is shown in a Ragone plot,* which displays measured values of specific power as a function of specific energy (see, for example, Figure 1 in the article by Nazri in this issue). A battery capable of delivering high current without a severe voltage penalty is said to possess good rate capability, or good ratability. The value of the cell current is often expressed as a so-called C rate, C/n, where C represents the total charge of the battery and n the time, in hours, required to drain the battery. This is a contentious unit, owing to the fact that the accessible capacity of the battery is itself a function of the drain rate, that is, $C = f(C/n)$. Cycling (i.e., repeated charging and discharging) generally leads to loss of capacity. Capacity fade is strongly dependent upon service conditions, particularly DOD and C rate.

Unsolved Problems

In large measure, enhancements in battery performance are throttled by limitations in materials. The improvements sought in the electrode materials include (1) higher capacity for lithium uptake, (2) higher lithium diffusivity for higher discharge current, (3) greater chemical and electrochemical stability, and (4) structural stability over a wider range of lithiation. The improvements sought in an electrolyte include (1) higher electrical conductivity for higher discharge current with lower Joule heating, (2) greater chemical and electrochemical stability in contact with the anode and cathode, and (3) single-ion conduction by Li^+ alone. Ideally, an electrolyte would have the electrical properties of a liquid and the mechanical properties of a solid. Such a material would serve both as electrolyte and separator and endow the battery with mechanical flexibility, a property unattainable in cells with conventional liquid electrolytes. Finally, there are so-called systems issues, most notably (1) the stability of the electrode–electrolyte interfaces and (2) the scalability of the synthesis and assembly processes. From the perspective of battery design, improving performance revolves around raising the fraction of the electrochemically active components (namely, the electrodes) to as close to unity as possible, while decreasing the fraction of the supporting components (e.g., the electrolyte, current collectors, and packaging) to as near zero as possible. Regarding this last point, reducing the thickness of the electrolyte and eliminating the need for binders and conductivity additives are desirable strategies. Materials research has an important role to play in attaining these goals.

Materials Research: Prospective Solutions

Within the broader framework of solid-state ionics, electrode materials and polymer electrolytes were treated in a previous issue of *MRS Bulletin*.[7] This time, the focus is much tighter: rechargeable lithium batteries.

It has been almost 30 years since ionic conductivity was observed in polymers[8] and almost 25 years since such materials were put forth as candidate solid electrolytes.[9] In the first article in this issue, Wright reviews recent developments and puts forth, based upon the Li^+ transport mechanism, a classification scheme that encompasses solvent-containing systems and true (solvent-free, or dry) solid polymer electrolytes, including novel "decoupled systems" exhibiting conductivities competitive with liquid electrolytes.

Simultaneously achieving high energy density and high power density (termed rate capability by battery designers, as mentioned earlier) necessarily requires a high electrode–electrolyte interfacial area coupled with short diffusion distances within the electrodes themselves. Clearly, there is a role here for nanotechnology. The article by Sides et al. describes how the authors have employed nanostructured electrodes prepared by template synthesis to achieve unprecedented rates of Li^+ movement through electrodes, translating into high power density and high energy

*The Ragone plot was introduced in a paper presented by D.V. Ragone at the Society of Automotive Engineers conference held in Detroit in May 1968.

density. Specific examples of nanofiber Sn-based anodes and nanostructured V_2O_5 cathodes are discussed.

In an effort to reduce costs by displacing $LiCoO_2$ from the cathode, layered $LiNiO_2$-based materials have been the subject of considerable attention. The article by Delmas and Croguennec details the synthesis, characterization, and electrochemical behavior of $LiNiO_2$ and describes how cationic substitution in this material can overcome its intrinsic performance limitations.

The characterization of materials found in lithium battery technology poses special challenges due to the presence of phases lacking long-range order and the low atomic number of Li. The article by Grey and Greenbaum shows the power of nuclear magnetic resonance in elucidating battery materials issues by probing local atomic environments and measuring the values of physical parameters important in battery operation. Among the latter are Li^+ mobility and changes of valence of the cathode-active cations.

Thanks to dramatic increases in computational power, computational modeling is playing an increasingly important role in materials research as well as in battery design. The article by Ceder et al. gives two snapshots of the field: one at the atomic level, where the focus is on the prediction of battery materials properties; and one at the systems level, where the focus is on the prediction of battery performance. The authors show how first-principles calculations enable the rapid screening of candidate materials and identification of the most promising candidates for subsequent laboratory study. En route, fundamental mechanisms can be elucidated, for example, the specific location of the compensating charge when Li^+ intercalates into the cathode, and the values of difficult-to-measure properties (e.g., calculating the concentration-dependence of lithium diffusivity), some of which are needed in macroscopic models. By simulating battery behavior under steady-state and transient conditions, macroscopic modeling serves to enhance the value of laboratory test results so as to decrease the length of the design cycle and lower the risk of innovation. Specific information includes predicting cell failure, identifying potential safety hazards, and even choosing the optimal cell geometry for a given application.

Implantable medical devices present a unique set of challenges for designers of portable power sources. This year marks the 30th anniversary of the first implant of a lithium battery in a human: a lithium/iodine cell that powered a cardiac pacemaker. The article by Takeuchi and Leising recounts the developments of primary (non-rechargeable) and secondary lithium cells since that time, drawing attention to the special requirements of batteries for human implantation. These include safety, reliability, predictability of performance, small size, and end-of-life indication, to name the most important. Materials advances in microelectronics and wireless communications are expected to enable radical innovation in this area, with an attendant demand for high-performance portable power.

To improve urban air quality, much attention has been given to so-called zero-emission vehicles, the primary example being the electric vehicle (EV). Everyone agrees that the weak link in the technology is the battery. Even the best commercially available rechargeable battery fails to meet consumer expectations for driving range. Clearly, without major improvements in battery technology, EVs will not gain widespread acceptance. Nazri explains the performance requirements for automotive traction. Interestingly, he makes the point that not only does EV technology stand to gain from better batteries, but it is likely that fuel-cell-powered vehicles will also require battery assistance. Also, with better batteries, the emissions from hybrid electric vehicles will decrease as they become less reliant on the engine for propulsion.

The Future

Niels Bohr, the Danish physicist and Nobel Laureate, once cautioned that prediction is always dangerous, especially when it is about the future. With this disclaimer, then, we speculate on what is in store for rechargeable lithium batteries. In the near term, expect the push for all-solid-state, flexible, thin-film batteries to continue. This is driven by the desire to maximize the electrode–electrolyte interfacial area while minimizing diffusion distances within the electrodes themselves, in order to combine high capacity with high rate capability. Recent results from our laboratory indicate that in a multilayer configuration comprising an anode of metallic lithium, a solid polymer electrolyte, and a cathode of dense, thin-film vanadium oxide, it is possible to construct a battery with projected values of specific energy exceeding 400 Wh/kg (700 Wh/l) and specific power exceeding 600 W/kg (1000 W/l).[10,11] Another trend is distributed power sources as opposed to a single central power supply. This allows for miniaturization (e.g., the microbattery). Expect also the integration of energy generation with energy storage, for example, a multilayer laminate comprising a photovoltaic charger and a rechargeable battery. Ultimately, if scientific discoveries prove to be scalable and cost-effective, we should witness the large-scale adoption of electric vehicles.

Acknowledgments

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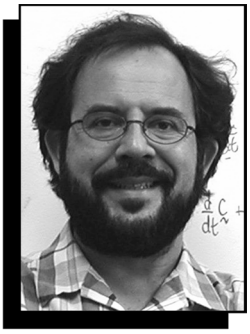

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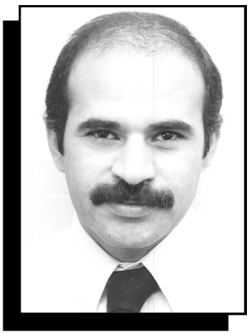

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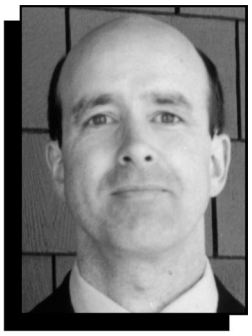

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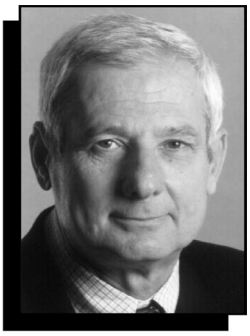

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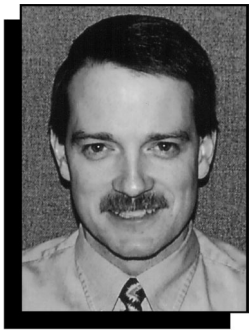

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