

# Batteries for Electric and Hybrid-Electric Vehicles

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## Key Words

anode materials, cathode materials, high-energy batteries

## Abstract

Batteries have powered vehicles for more than a century, but recent advances, especially in lithium-ion (Li-ion) batteries, are bringing a new generation of electric-powered vehicles to the market. Key barriers to progress include system cost and lifetime, and derive from the difficulty of making a high-energy, high-power, and reversible electrochemical system. Indeed, although humans produce many mechanical and electrical systems, the number of reversible electrochemical systems is very limited. System costs may be brought down by using cathode materials less expensive than those presently employed (e.g., sulfur or air), but reversibility will remain a key challenge. Continued improvements in the ability to synthesize and characterize materials at desired length scales, as well as to use computations to predict new structures and their properties, are facilitating the development of a better understanding and improved systems. Battery research is a fascinating area for development as well as a key enabler for future technologies, including advanced transportation systems with minimal environmental impact.

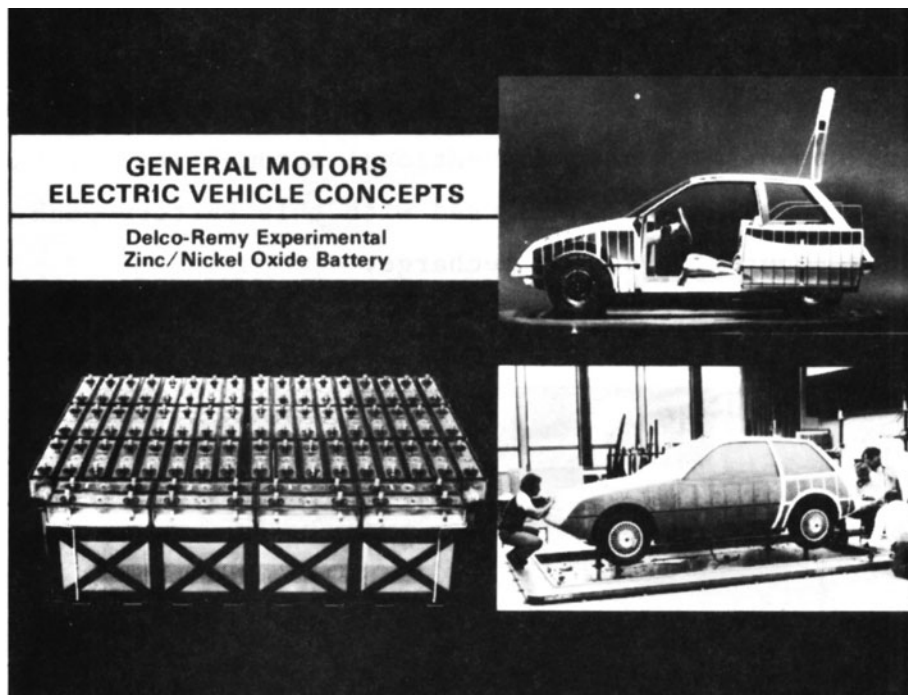
## INTRODUCTION

Batteries have been in use in vehicles for more than a century. Therefore, there is a widespread feeling that battery technology is stagnant, and indeed, only significant advances will enable electric vehicles (EVs) to meet consumer acceptance. Throughout this review we discuss the principal problems faced by battery designers and some of their methods to overcome those problems. Although significant progress has been made, the problem lies more with the demanding applications battery designers are pursuing. Batteries are among the very few reversible electrochemical systems in mass production today and therefore have unique challenges. We also include a brief history of battery-powered vehicles and basic information on battery components and principles of operation.

## HISTORY

EVs have a long history dating back to approximately 1834, when Robert Anderson of Scotland built the first simple EV (1). At that time, because there were no rechargeable batteries, the battery had to be replaced after each full discharge. It was not until 1859, with the invention of the rechargeable lead-acid battery by Gaston Plante of France, that the electric car became more practical. In the late 1800s, there was significant interest in electric automobiles, and competition developed concerning the best battery for EVs. Just before the dawn of the 20th century, more automobiles were EVs than were gasoline powered. A large number of small companies produced these electric automobiles using hand-assembly methods. Thomas Edison entered the competition for a better EV battery with his development of the Fe/NiOOH battery containing an alkaline electrolyte, which was first marketed in 1904 and then reintroduced with an improved design in 1908. These batteries were very robust, some of them lasting for a few decades. With Henry Ford's invention of mass-production methods for automobiles in 1910, the gasoline-fueled vehicle became affordable for many people, and the electric automobile was gradually displaced from the market because of relatively high cost and limited range. In 1912 Charles Kettering invented the electric starter for gasoline vehicles. Since then, the main automotive application for batteries has been for starting, lighting, and ignition. From 1930–1960 there was little activity in battery-powered EVs.

The oil embargo of 1973 caused a rapid rise in the price of oil and gasoline as well as supply shortages. The long lines at gasoline stations and competition for fuel enhanced interest in reducing our dependency on oil. Renewed interest in electric automobiles was thus born. The best lead-acid EV technology available in the 1970s yielded a range of less than 50 miles. For example, the Vanguard-Sebring CitiCar, introduced to the EV market in 1974, operated on Pb/PbO<sub>2</sub> batteries. It could cruise at 30 mph and had a range of approximately 40 miles. Not many were sold, and the CitiCar was withdrawn from the market after only a couple of years. The limited range of these EVs made them unattractive to consumers. In the 1970s General Motors (GM) undertook a program to develop electric automobiles and pickup trucks using Zn/NiOOH batteries (developed by GM) for improved performance and range (2, 3). The Zn/NiOOH battery pack and the modified Chevette (Electrovette) are shown in **Figure 1**. These vehicles had an Environmental Protection Agency (EPA) urban range of 60 miles, a range of 80 miles at a constant speed of 45 mph (4), and acceleration similar to that of gasoline-powered vehicles. The life of the test batteries was approximately 41,000 miles (5). The Electrovette and the Chevy Luv Truck EVs were planned for market entry, but when the oil embargo ended and gasoline prices and availability improved, the EV production plan was abandoned. In addition, during the 1970s there was significant interest in the development of batteries that operated at elevated temperatures and delivered much higher



**Figure 1**

General Motors Zn/NiOOH battery pack and Electrovette model, which were developed in the 1970s.

specific energy than ambient-temperature batteries. The main batteries of this type were the Na/S and Li/FeS<sub>2</sub>, each of which operated at 350–400°C (6–8). These never reached production for EVs, but a small number of demonstration vehicles were built.

The 1990s saw a renewal of interest in EVs that was driven largely by a California Air Resources Board mandate that the seven major manufacturers selling vehicles in California have 2% of their sales in 1998 and 10% of their sales in 2003 be vehicles with zero emissions (9). Under industry pressure due to concerns about consumer interest in EVs, in 1996 the mandate for selling EVs in 1998 was pushed back, and by 2000 the requirements for the sale of fully electric vehicles were replaced by requirements for other types of clean cars (10, 11). A well-known product of this period was GM's EV1. The first generation used a lead-acid battery and had a range of 80–100 miles, whereas the second generation used a nickel-metal hydride (Ni/MH) battery and had a range of 100–140 miles. GM ultimately canceled its leasing program and destroyed most of the vehicles because the company could not sell enough to make the program financially viable. Indeed, demand was weak while the EV1 was available for lease (12).

The 2000s saw a renewed interest in EVs by manufacturers and consumers owing not to government mandates but to high oil prices and technological developments in lithium-ion batteries that made their performance and cost increasingly attractive. The performance of lithium-ion batteries improved such that the specific energy of a commercial cell reached 200 Wh kg<sup>-1</sup>, more than twice that of a Ni/MH cell. In addition, the price of a lithium-ion (Li-ion) cell fell below that of a Ni/MH cell (13). Based on the improvement in battery performance and decrease in cost, Tesla Motors was founded in 2003 to build a high-performance EV. By 2009 it was delivering a high-performance sports car and had plans to build a sedan (14, 15). It is currently the only serial

manufacturer of EVs in North America or Europe, although most major automakers now have plug-in hybrid-electric and all-electric-vehicle programs, and some have plans to release a vehicle within the next few years. Essentially all major EV programs currently plan to use lithium-ion cells.

## BATTERY BASICS

A battery is made up of electrochemical cells. Each cell has a negative electrode, a positive electrode, and an electronically insulating but ionically conductive layer separating the electrodes (16–20). Electronic current passing through an external circuit can do work on a device (such as an electric motor). Batteries for EVs must also be rechargeable, which implies that the electrochemical reactions that occur at each electrode must be reversible over thousands of cycles. To illustrate the basic battery components, a cross-sectional image of a lithium-ion cell is shown in **Figure 2**. Each electrode is composed of an active material at which electrochemical reactions occur as well as various additives that often include a material (such as carbon black or other forms of carbon) to improve the electronic conductivity of the electrode and a polymeric binder to hold the particles and the conductive additive together. For the lithium-ion system shown in **Figure 2**, the positive-electrode active material is a metal oxide such as  $\text{Li}_y\text{CoO}_2$ , and the negative-electrode active material is  $\text{Li}_x\text{C}_6$ . The current collectors provide a low-resistance electronic pathway to the external circuit and must be chosen to avoid corrosion reactions.

As shown in **Figure 2**, the electrodes are porous structures, and the electrolyte is present throughout the electrodes as well as in the separator. A porous electrode increases the surface area for the electrochemical reaction and reduces the diffusion distances within the active materials. Indeed, it offers advantages similar to those of a packed bed reactor. The electrode thicknesses are chosen to minimize the amount of electrochemically inert material while ensuring that the active material is accessible for a characteristic discharge time that depends on the application (17). Various methods have been devised for optimizing electrode thicknesses (22, 23).

The design of a reversible chemical system is very difficult. From an essentially infinite set of possible electrode combinations, only a small number are sufficiently reversible to be of commercial interest. Intercalation reactions are an important class of reversible reactions and occur in the Ni/MH and most lithium-ion systems. Intercalation reactions involve the reversible insertion and extraction of an atom into a crystal lattice with minimal changes to the host crystal during the intercalation process (for example, the volume change is typically less than 10% during a complete insertion or extraction process) (24, 25). The dominant lithium-ion chemistry today involves the intercalation of lithium at each electrode. At the positive electrode, the reaction (for a  $\text{LiCoO}_2$  electrode) is

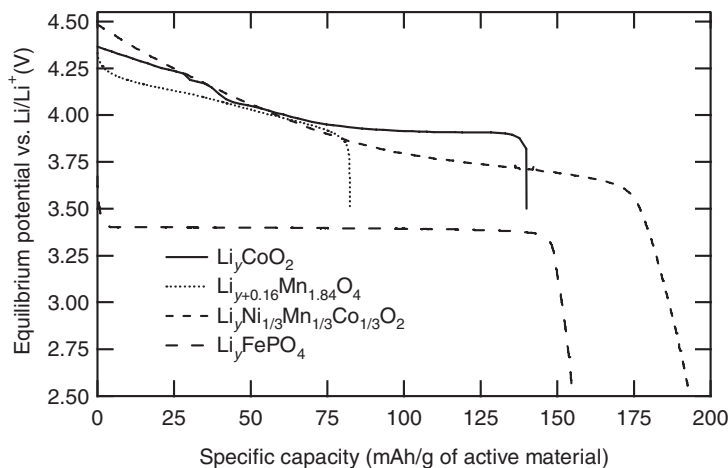


and at the negative electrode (for a carbon electrode) the reaction is



There are reversible systems based on other types of reactions. For example, the lead-acid battery uses a dissolution-precipitation mechanism with the reaction product,  $\text{PbSO}_4$ , sparingly soluble in the concentrated  $\text{H}_2\text{SO}_4$  electrolyte. Other types of electrodes include those with metals that anodically dissolve and cathodically redeposit (such as Zn), and those involving the formation of alloys (such as Li in Al) (26, 27).

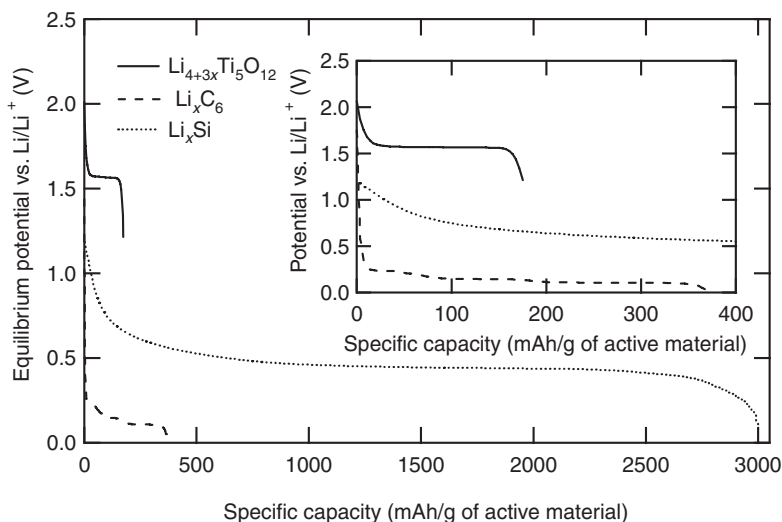
The potential at which an electrochemical reaction occurs is related to the energy levels of the products and reactants. Although atomistic simulations have made some progress in predicting



**Figure 3**

The equilibrium potential of the reactions at several electrodes commonly used as positive electrodes in lithium-ion systems (22, 32–34). Although each material intercalates Li, the magnitudes and shapes of the equilibrium potential curves differ significantly.

energy levels (28–30), in general the nonideal thermodynamics of the materials necessitate direct measurement. **Figure 3** shows the potential at which a variety of reactions at the positive electrode of a lithium-ion cell occur, and **Figure 4** shows reactions at the negative electrode. Clearly, different materials can have very different equilibrium thermodynamics. The figures also show the variation in the amount of charge that can be stored per gram of active material. Although not always true, a high specific capacity often corresponds to a significant amount of volume expansion and contraction during cycling. For example, Li forms an alloy with Si at compositions



**Figure 4**

The equilibrium potentials of the reactions at several electrodes commonly used as negative electrodes in lithium-ion systems.

up to  $\text{Li}_{4.4}\text{Si}$ , and over the full range of alloy composition, the volume changes by approximately a factor of three (31). Materials with significant volume changes during cycling experience greater mechanical stresses, especially at high current densities, and therefore may be more likely to fracture and degrade (24, 25).

An electrochemical cell involves coupled reactions at two electrodes, and the potential of the cell is calculated by subtracting the potential at the negative electrode from the potential at the positive electrode. From **Figures 3** and **4** we can see that the potential of a lithium-ion cell with a  $\text{Li}_y\text{CoO}_2$  positive electrode and a  $\text{Li}_x\text{C}_6$  negative electrode is approximately 3.85 V, although it varies during the course of a charge or discharge.

The theoretical specific energy of a cell (in  $\text{Wh kg}^{-1}$  of active material) is given by

$$\hat{E} = \frac{U_{pos} - U_{neg}}{\frac{1}{\hat{C}_{pos}} + \frac{1}{\hat{C}_{neg}}}, \quad 3.$$

where  $U$  is the average equilibrium potential (in V), and  $\hat{C}$  is the specific capacity (in  $\text{Ah kg}^{-1}$ ). The presence of the electrochemically inactive materials in the cell can be included easily to estimate the specific energy of a practical cell, which is usually less than half of the specific energy based on the active materials alone. The energy per unit volume can be calculated if the densities of the cell components are known. **Table 1** gives the theoretical specific energy (based on the weight of active materials alone) and other properties of several battery chemistries.

Although the specific energy can be calculated easily, there are no simple equations to find the power produced by a cell. The potentials shown in **Figures 3** and **4** are equilibrium potentials, but when operated at finite rates, impedance in the cell causes the operating potential to differ from the equilibrium potential. Important sources of impedance in a cell include ohmic resistance (resistance to the passage of electronic or ionic current through a given medium), kinetic resistance (resistance to the transfer of current across an interface), and mass-transfer resistance (the buildup of concentration gradients in the active materials and the electrolyte). The importance of each of these resistances depends on the materials, the battery design, and the method of operation. For

**Table 1** Specifications of conventional and advanced battery chemistries. If a metal-air cell is open to the environment, its weight will increase during discharge. Theoretical specific energies are calculated using Equation 3

		Theoretical specific energy ( $\text{Wh kg}^{-1}$ )		Specific energy of practical cell ( $\text{Wh kg}^{-1}$ )	Cell potential (V)	References
Conventional cells						
	$\text{C}_6/\text{LiCoO}_2$	584		220	3.80	(35, 36)
	MH/NiOOH	195		70	1.2	(37–42)
	Pb/PbO <sub>2</sub>	240		35	2.0	(16, 43, 44)
Advanced cells: Li/S						
	Li/S	2564		350	2.2	(45, 46)
Advanced cells: metal/air						
Metal	Discharge product	Metal only	Discharge product			
Zn	ZnO	1312	1054	350	1.6	(16)
Al	$\text{Al(OH)}_3$	8047	2783	250	2.7	(16)
Li, nonaqueous	$\text{Li}_2\text{O}_2$	11970	3622		3.1	(16)

example, if a battery receives primarily short pulses with intermediate rest periods, the buildup of concentration gradients may be minor. For a state-of-the-art  $\text{Li}_x\text{C}_6/\text{Li}_y\text{CoO}_2$  cell, the area-specific resistance is typically approximately  $20 \text{ ohm}\cdot\text{cm}^2$ , and the dominant sources of resistance in the cell include kinetic resistance to charge transfer across the interfaces and a resistive film that forms on the surface of the  $\text{Li}_x\text{C}_6$ . For sustained high currents, mass-transfer resistance in each active material and the electrolyte become important.

The performances of various systems are compared with a Ragone plot, which gives the specific energy and specific power for different discharge rates, as shown in **Figure 5**. The ordinate shows the specific energy, and the abscissa gives the specific power. The diagonal lines correspond to different discharge times, with long discharge times to the upper left. As discussed above, at a high discharge rate, less energy can be extracted because of the increasing magnitude of losses owing to the resistances within the cell. In addition, the resistances are nonlinear, which accounts for the rapid drop in specific energy at very high specific powers. **Figure 5** shows results for the three main types of rechargeable cells on the market today, the lead-acid, Ni/MH, and lithium-ion systems. The lithium-ion system has the best performance, although the Ni/MH system can also meet the U.S. Department of Energy (DOE) goal for the hybrid-electric vehicle (HEV) application (DOE goals are discussed in the next section). To provide a fair comparison between the performances of different chemistries, an optimized Ragone plot should be constructed with the cell design adjusted for each discharge time (22, 23, 38).

## U.S. DEPARTMENT OF ENERGY GOALS FOR BATTERY PERFORMANCE FOR ELECTRIC-POWERED VEHICLES

The DOE has set goals for battery-system performance, which are summarized in **Table 2** (47–49) and reflect the DOE assessment of the system-level attributes necessary for a battery in a particular application to become competitive with current vehicle technology. Goals exist for three designs: hybrid-electric, plug-in hybrid-electric, and electric vehicles. For reference, a state-of-the-art lithium-ion electric-vehicle pack has, at the system level, a specific energy of approximately  $120 \text{ Wh kg}^{-1}$  and a specific power of approximately  $450 \text{ W kg}^{-1}$  [these numbers are for the Tesla Roadster battery pack, which weighs 450 kg, has an energy of 53 kWh, and uses approximately

**Table 2** DOE goals for system-level performance for three electric-powered vehicle types. The min and max categories refer to designs that meet the basic HEV or PHEV application but have a minimum or maximum amount of battery. The EV numbers are the short-term ones given by the DOE

Performance criteria	HEV-min	HEV-max	PHEV-min	PHEV-max	EV-short term
Specific power, $\text{W kg}^{-1}$	625	667	750	316	300
Specific energy, $\text{Wh kg}^{-1}$	7.5	8.3	57	97	150
Power density, $\text{W L}^{-1}$	782	889	1125	475	460
Energy density, $\text{Wh L}^{-1}$	9	11	90	145	230
Pack energy, kWh	0.3	0.5	3.4	11.6	40
Pack weight, kg	40	60	60	120	267
Pack volume, L	32	45	40	80	174
Life, years	15	15	15	15	10
Cycle life	300,000	300,000	5000	5000	1000
System price, \$	500	800	1700	3400	6000

HEV: hybrid-electric vehicle, PHEV: plug-in hybrid-electric vehicle, EV: electric vehicle.



6800 cells of the 18650 size (similar to AA size)] (50). In addition to the requirements in **Table 2**, the battery system must be able to operate within a temperature range of  $-40$  to  $+50^{\circ}\text{C}$ . In going from the cell to system level, the mass significantly increases to account for the mechanical support, electrical interconnects, controls, and cooling system. It is thus necessary to compare performance numbers on a common weight basis because results for a single cell are often approximately 50% more than those for that same cell in a pack.

Current battery systems fall short of the DOE goals principally in life and system price. For example, the DOE EV goals have a system life target of 10 years, but it remains unclear how long present batteries can last in field use because only accelerated-aging tests have been done. The life of a battery often falls significantly with increases in operating temperature; therefore, batteries in warm climates may have a shorter life than those in cooler climates. System cost remains a major challenge and is also tied into life because if a battery pack needs to be replaced, for example, every five years, two or three packs will be needed during the vehicle's life. Although estimates are difficult, a present pack-level cost of approximately  $\$500\text{--}700\text{ kWh}^{-1}$  is reasonable; the 40-kWh pack specified in the DOE goals for an EV thus would cost at least  $\$20,000$ , more than a factor of three too high.

What accounts for the high cost? Argonne National Laboratory found that for a high-energy lithium-ion cell, the major contributors to cost were the cathode-active material ( $\sim 49\%$ ), electrolyte ( $\sim 23\%$ ), and anode-active material ( $\sim 11\%$ ) (51). The prices of the raw materials used to make the cathode-active materials, such as Co and Ni, may fluctuate and change the final cell price. Overall, approximately 70% of the cost of a lithium-ion cell is due to materials, with the remainder manufacturing and other costs.

## BATTERY DESIGN CONSIDERATIONS

### Driving Profile

Driving profiles are important for the engineering of all types of vehicles, including EVs. The driving profile represents the velocity versus time relationship for a typical vehicle of a certain type used in a specified manner. For example, highway driving would be represented by a reasonably constant velocity, perhaps averaging 60 mph, with occasional higher and lower speeds. An urban driving profile would be characterized by frequent stops and starts and a velocity averaging 20–30 mph. An example of an urban driving profile is the Federal Urban Driving Schedule, shown in **Figure 6** (52). These driving profiles are important in vehicle design because they determine power and energy requirements. The driving profiles in combination with the equations of motion for the vehicle provide a power profile that the battery must meet. The peak power required for acceleration is typically 7–10 times the average power for urban-suburban driving (53).

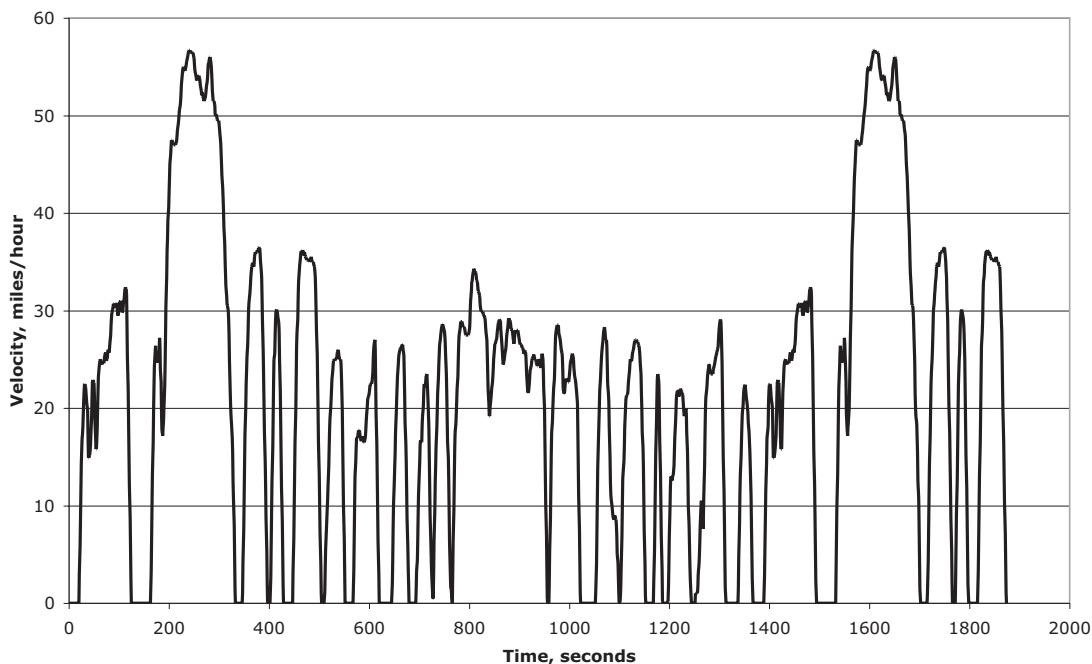
### Cell Design

Commercial electrochemical cells are typically produced in the cylindrical format, as in common flashlight cells, and the prismatic format, as in cells in an automotive lead-acid battery or many cell phones and digital cameras (**Figure 7**). These two formats are produced in a wide range of sizes, from cells measuring a few centimeters on a side to cells measuring tens of centimeters on a side. The cylindrical format cell has a manufacturing advantage in its smaller parts count but has a disadvantage with respect to space utilization.

Current produced in a cell must be gathered for flow to or from an external circuit, and a metal such as Cu or Al is often used. The placement of the tab influences the current distribution within a



## Federal Urban Driving Schedule



**Figure 6**

The Federal Urban Driving Schedule (FUDS) shows the variable velocity a vehicle power system must provide.

cell because it determines the resistance of the current pathways. In general, active material closer to the tab experiences a higher current density. The tabs, with their large amount of current, are also an important site of heat generation. The National Renewable Energy Laboratory has taken some images that show how the introduction of an additional pathway for current affected the surface temperature of a Prius module when it went through a redesign (54). An optimization that takes into account both performance and cost needs to determine the placement and sizing of the tabs.

## Thermal Management

Thermal management is a crucial aspect of cell and pack design and a major reason for the significant expense of high-energy packs; it is needed to maximize performance, life, and safety (55–61). It affects performance because many physical processes in a battery depend on temperature (for example, the internal resistance of the battery typically falls with increasing temperature). However, because the rate of battery aging increases with rising temperature, typically an optimal temperature regime exists (approximately 30–40°C). A basic goal for a thermal management system is to keep all battery regions at roughly the same temperature and below the value at which aging becomes rapid (60, 62, 63). Large battery packs in EVs thus require a liquid cooling system connected to radiators, whereas for the smaller batteries used in HEVs, air cooling (possibly using conditioned air from the cabin) is adequate.

A thermal management system is also required to ensure that if a cell goes into thermal runaway (e.g., because of an internal short circuit), it will not propagate to other cells. If a propagation of thermal runaway from cell to cell begins, a catastrophic failure is possible. A thermal runaway

involves the initiation of exothermic reactions in the cell (e.g., in lithium-ion cells the organic electrolyte can combust) and possibly the melting of cell components. The potential for a catastrophic failure in a large battery pack may be higher than that within a gasoline-powered vehicle because in the latter the oxidant (air) is separate from the reductant (gasoline), whereas in the former the oxidant and reductant are in close proximity. In the case of lithium-ion cells, the separator thickness is often just 25  $\mu\text{m}$ .

There is a tradeoff between many small cells, which have a high surface-to-volume ratio and allow relatively easy cooling, but require a large number of electrical interconnects, and large cells, which have a low surface-to-volume ratio and make cooling difficult, but require a small number of interconnects. It remains to be seen where the optimal cell number and size lie. It should also be noted that prismatic cells may have a more uniform temperature distribution and allow easier thermal management because they can be stacked.

## VEHICLE BATTERIES IN CURRENT USE

### The Ni/MH System

The dominant system in use today for HEVs is the Ni/MH chemistry (38, 64–66). The Ni electrode has been used for many decades and was paired with a hydrogen, iron, cadmium, or zinc electrode prior to a MH electrode, which was developed more recently (16). The Ni/MH system itself dates from the 1960s. The first consumer cell was sold in the late 1980s, and Ni/MH batteries became available in cars in the EV1 and later as the chemistry of choice for HEVs. Recently, Toyota announced that it plans to continue using Ni/MH batteries in its HEVs, solidifying the importance of Ni/MH batteries for years to come (67).

The Ni/MH cell has a  $\text{Ni}(\text{OH}_{1-y})_2$  positive electrode. The material intercalates protons, has a theoretical specific capacity of 289  $\text{mAh g}^{-1}$ , and has a potential that exhibits an interesting hysteresis between charge and discharge that makes state of charge (SOC) determination difficult (42, 68). The  $\text{Ni}(\text{OH}_{1-y})_2$  electrode is constructed from a Ni foam that is a porous current collector framework of Ni metal onto which the  $\text{Ni}(\text{OH}_{1-y})_2$  is deposited. The negative electrode is a metal-hydride material. The MH electrode can contain a range of elements, although there are two main classes of materials, termed  $\text{AB}_2$  and  $\text{AB}_5$  (39). A characteristic  $\text{AB}_5$  material used in MH electrodes is  $\text{LaNi}_5$ , although other elements are often included (69–71). The MH electrode typically exists in two phases over most of the discharge range, resulting in a flat potential. The electrolyte is a concentrated (approximately 7 M) aqueous solution of KOH. Relative to the electrolytes used in lithium-ion cells, the transport properties are extremely good, allowing thick separators (e.g., the separator in the 2005 model year Prius battery is 220  $\mu\text{m}$  thick) to be used even for cells intended for high-power applications. Detailed performance plots for the Ni/MH system have been published (38, 72).

A major benefit of the Ni/MH system is the presence of side reactions that provide a measure of overcharge protection and cell balancing. At the potentials present at the end of a charge at the positive electrode, oxygen may be evolved; the oxygen can diffuse across the cell to the negative electrode, where it is recombined (reduced). At low rates (up to a current that would result in a full charge or discharge in 10 hours, although this depends on cell design), this oxygen shuttle can carry all the current that is passed to the cell. This shuttle also helps with capacity balancing. The lack of such a shuttle in other chemistries, such as lithium-ion, causes significant problems.

The Ni/MH chemistry is relatively safe because it uses an aqueous electrolyte rather than an organic solvent that could ignite if a short circuit occurs. However, gas generation can occur

during normal operation, and Ni/MH cells are typically built with a one-way gas vent to allow hydrogen, oxygen, or water vapor to escape.

Evidence points to an excellent lifetime for the Ni/MH system. The Ni electrode has been shown to have an extremely long life. Major problems include degradation of the MH electrode and loss of water from the electrolyte that causes the cells to dry out. The dominant failure mechanism in lithium-ion cells—side reactions that shift the state of charge balance and reduce the usable capacity of the cell—is not present in Ni/MH cells (73).

Although on the cell level the cost of a lithium-ion cell per kWh of energy stored may now be lower than that of a Ni/MH cell, for a vehicle application the relevant cost comparison is the pack. The better inherent safety and long life of the Ni/MH chemistry mean that it is still less expensive than lithium-ion at the pack level (13, 67).

## Lithium-Ion Systems

Lithium-ion cells, introduced commercially in 1991 by the Sony Company, quickly captured the market for batteries in camcorders, cell phones, and cameras (35, 36). This was possible because of the high specific energy (more than 100 Wh kg<sup>-1</sup>) and long cycle life (more than 1000 deep cycles) of Li-ion cells. These cells were very expensive (initially, more than \$20,000/kWh), and could be used only in applications that would tolerate this cost. Li-ion cells use carbon as a host structure for lithium at the negative electrode and cobalt oxide as the positive electrode active material. The electrolyte is a mixture of organic solvents and a lithium salt. The cells are hermetically sealed to prevent ingress of air or water, which would react with the electrodes. **Figure 7a** shows a drawing of a spirally-wound cylindrical Li-ion cell. The electrodes are very thin (typically 20–50 μm but as much as 100 μm for some applications), and have a large area, allowing them to operate at low current densities relative to those used in aqueous electrolyte cells. This is important because the organic solvent-based electrolytes have relatively low conductivities, and the diffusion of the Li into the positive-electrode active material is slow.

Soon after the Li-ion cells were introduced, it was recognized that the cobalt oxide used in the positive electrode is quite expensive and not environmentally benign. Safety soon emerged as another important issue because overcharge, overdischarge, or overheating could cause the cells to burst into flame. Significant research and development has been devoted to solving these problems. Researchers have investigated positive electrode materials to reduce cost and improve safety, negative-electrode active materials to reduce the weight, and electrolytes to extend the temperature range of safe operation, improve the conductivity, and reduce the reactivity while maintaining a stable, protective film (the solid-electrolyte interphase) on the negative electrode.

Some of the many positive-electrode active materials that have been investigated are listed in **Table 3**. The positive-electrode active materials used in lithium-ion cells all have structures that are sufficiently open to allow the insertion and removal of lithium ions, either in tunnels or between atomic planes of the structure, with minimal disruption of the crystal structure (74, 78). The first commercially successful lithium-ion cells used LiCoO<sub>2</sub>, which has a layered structure that accommodates the insertion of Li between the CoO<sub>2</sub> planes (36). Unfortunately, the removal of too much Li results in a phase change that causes performance loss. Because of this restriction, only approximately half of the lithium can be removed while retaining capacity with extended cycling. Under these conditions, approximately 140 mAh g<sup>-1</sup> specific capacity can be used with a cycle life of more than 1000 cycles (35). This is excellent cycle life, and the current specific energy of approximately 200 Wh kg<sup>-1</sup> permits many applications in the field of portable electronics. The safety of the Li<sub>x</sub>C<sub>6</sub>/Li<sub>y</sub>CoO<sub>2</sub> cell has been problematic, and measures taken to improve safety

**Table 3** Specifications of selected positive-electrode materials

Material	Structure	Practical specific capacity (mAh g <sup>-1</sup> )	Cycle life	Comments	References
LiCoO <sub>2</sub>	Layered	140	>1000	Safety issues, expensive	(35, 36, 74)
LiNiO <sub>2</sub> -based	Layered	140	300	Safety issues, stability	(74–76)
LiMn <sub>2</sub> O <sub>4</sub>	Spinel	120	300	Spinel structure not stable with cycling	(74, 77–80)
LiMn <sub>1/3</sub> Ni <sub>1/3</sub> Co <sub>1/3</sub> O <sub>2</sub>	Layered	150	300	Relatively high specific energy	(74, 77, 78, 81)
LiFePO <sub>4</sub>	Olivine	150	>1000	Safe, can have high specific power	(74, 77, 78, 82, 83)

include protective microcircuits on each cell and battery that prevent overcharge and overdischarge. The cost of Li<sub>x</sub>C<sub>6</sub>/Li<sub>y</sub>CoO<sub>2</sub> cells has gradually decreased but remains too high for consumer EV or HEV applications.

To reduce cost and improve safety, various other cathode materials have been investigated, as shown in **Table 3**. Attempts to eliminate Co have not been successful for the layered oxide materials. In the case of LiNiO<sub>2</sub>, some of the Ni occupies sites in the Li plane, interfering with Li movement and reducing the capacity (76). Adding some Co results in higher capacity and less Ni on Li sites (74). The LiNiO<sub>2</sub>-based materials also have significant safety problems due to the catalytic activity of the Ni. As a result, these materials are not commercially attractive.

Manganese-based materials initially offered hope for developing a low-cost cathode material that is environmentally benign. The LiMn<sub>2</sub>O<sub>4</sub> spinel material was investigated intensively for several years but suffered from the conversion of the spinel structure to orthorhombic, with consequent capacity and performance loss (74). The cycle life can be improved by maintaining the Li content below one Li per two Mn. If more Li is inserted, the structure changes irreversibly. Another issue is the disproportionation of the Mn<sup>+3</sup> to Mn<sup>+2</sup> and Mn<sup>+4</sup>. Mn<sup>+2</sup> is soluble in the electrolyte and can migrate away from the cathode, resulting in capacity loss. Attempts to solve these problems have not been very successful, although doping with such elements as Al, Zn, or Cr does result in some improvement (84).

The next stage in the development of the transition metal oxide materials was the use of combinations of Mn, Ni, and Co in varying amounts in the layered oxide structure (85–87). In the development of these materials, the Co content is minimized, and the Mn content is maximized while attempting to maintain high specific capacity and rate capability as well as good safety. In some cases, dopants such as Al or Fe are added (88). In general, good specific capacity and rate capability are maintained, but the cost is still high for large consumer batteries.

The most recent commercial entry in the rechargeable Li battery field is the Li<sub>x</sub>C<sub>6</sub>/Li<sub>y</sub>FePO<sub>4</sub> cell. The cathode material is inherently low cost, easy to synthesize, quite safe, and environmentally benign. The cell voltage (3.4 V) is lower than that of the oxide-based cells (3.5–4.0 V). Early work on LiFePO<sub>4</sub> cathodes indicated a lower rate capability because of the low electronic conductivity of the LiFePO<sub>4</sub>. There are many reports of attempts to improve the electronic conductivity through the use of carbon in intimate contact with the LiFePO<sub>4</sub> and the synthesis of smaller particles of LiFePO<sub>4</sub>. The best-performing material now uses dopants to improve electronic conductivity and nanosized particles to reduce the diffusion distance of Li in the material. This improved material can be discharged in about one minute. Two companies have commercialized the Li<sub>x</sub>C<sub>6</sub>/LiFePO<sub>4</sub> cell (89, 90), which is gradually penetrating the market for power tools and is being used in demonstration electric and hybrid-electric vehicles. Other transition metal phosphates

**Table 4 Properties of anode material for lithium-ion cells**

Material	Theor. sp. capacity mAh g <sup>-1</sup>	Voltage versus Li	References	Comments
Li	3861	0.0	(94)	Safety issues with organic solvents
Li <sub>4.4</sub> Si	2012	0.45	(102–104)	Short cycle life
Li <sub>4.4</sub> Sn	790	0.5	(105)	Short cycle life
LiC <sub>6</sub>	350	0.25	(93)	Long cycle life
Li <sub>4</sub> Ti <sub>5</sub> O <sub>12</sub>	233	1.5	(106)	Long cycle life
Li <sub>2</sub> MgSn	340	0.4	(107)	Short cycle life

such as MnPO<sub>4</sub>, NiPO<sub>4</sub>, and CoPO<sub>4</sub> have also been explored, albeit with little success thus far (91, 92).

The negative electrode used in Li-ion cells is usually some form of carbon. Typically these carbon anodes can provide 350 mAh g<sup>-1</sup> specific capacity (93, 94). During the past several years there has been an effort to increase the specific capacity of the anode by using other host materials including silicon, tin, aluminum, and various intermetallics (31, 95, 96). Efforts also continue on the use of Li metal as a negative electrode (97). The theoretical specific capacities of some anode materials are shown in **Table 4**. The longest cycle lives are reported for the carbon materials and Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub>, with more than 1000 cycles. The other materials in **Table 4** have much shorter cycle lives owing to the large volume changes caused by Li insertion/removal. The large volume changes cause fracturing of the particles and loss of electronic contact, which results in capacity loss. All commercially available Li-ion cells contain carbon-based negative electrodes, though a few do have some Sn added. If the Li metal electrode could provide the necessary cycle life, then the specific capacity of the negative electrode would be dramatically improved, and the specific energy would increase as well.

## RAW MATERIALS SUPPLIES

If EVs were to be produced in large numbers, the issue of material supply comes into focus as a possible concern. Lithium, cobalt, and nickel are key materials for HEV and EV applications. The world production of lithium (as Li<sub>2</sub>CO<sub>3</sub>) in 2008 was 27,400 metric tons, and the reserves are estimated as 13 × 10<sup>6</sup> tons (98). This corresponds to the production of 5 × 10<sup>6</sup> vehicles per year, each with a 40 kWh battery. This is only a small fraction of the existing production rate of automobiles. If all of the lithium reserves were to be used in EVs, then the total produced would be approximately 2.4 × 10<sup>9</sup> vehicles. Clearly, production rates of lithium must be increased. The reserves are probably adequate.

The cobalt supply is much smaller. It is estimated that the world production for 2010 will be approximately 80,300 metric tons, with reserves of 4 × 10<sup>6</sup> tons (99). If this cobalt is used in LiC<sub>6</sub>/LiCoO<sub>2</sub> batteries of 40 kWh each for EVs, then 1.8 × 10<sup>6</sup> vehicles per year could be produced. The total world reserves could provide approximately 89 × 10<sup>6</sup> vehicles. Even without the high cost of Co, it is necessary to drastically reduce or eliminate the Co from EV batteries if large production rates are to be achieved.

The outlook for nickel is very good compared to that of lithium or cobalt. The world production of Ni is 1.57 × 10<sup>6</sup> metric tons, and the reserves are approximately 1.6 × 10<sup>8</sup> tons (100, 101). The current production rate of Ni corresponds to 36 × 10<sup>6</sup> vehicles per year, each with a 40 kWh LiC<sub>6</sub>/LiNiO<sub>2</sub> battery.

## CANDIDATES FOR BATTERIES OF THE FUTURE

Research continues on electrochemical couples that may serve as batteries in the future (108–111). The materials investigated depend, of course, on the application. Whereas specific energy and power are not important for stationary applications (cost being the main driver), for vehicle applications they are of primary concern. However, even within vehicle applications, the battery materials of the future for a HEV are likely to be significantly different than those for an EV. HEV batteries require materials that have very high rate capability and very good cycle life, such as the  $\text{Li}_{4+3x}\text{Ti}_5\text{O}_{12}$  negative electrode and  $\text{Li}_y\text{FePO}_4$  positive electrode (23, 112–114). However, for an EV in which the available range on a single charge remains a major limiting factor, specific energy is the main barrier. Another barrier is cost, which may scale with the total battery mass. Because a battery technology has already been established for HEV markets (the Ni/MH system), systems with a higher specific energy are a particular focus of interest and research and thus our focus in this section.

### Metal/Air Cells

Metal/air cells can offer, at least theoretically, very high specific energies. **Table 1** shows the specific energies of a select group of metal/air chemistries. Whereas some authors report the theoretical specific energy based on the weight of the metal alone, during discharge oxygen is stored in the battery, so it is equally appropriate to base the theoretical specific energy on the final discharge product. We report both in **Table 1**.

There are currently two major problems with metal/air cells. First, they exhibit a large polarization (internal resistance) upon charge and discharge even at low current densities, and second, they exhibit a very poor cycle life when recharged electrically. The high polarization at low current densities is related to the air electrode, which is well known to have poor kinetics. Even after decades of research, no catalyst has been found to significantly reduce the polarization, even under open-circuit conditions (115). The polarization limits the energy efficiency on charge and discharge to a value significantly lower than that achieved by present lithium-ion cells (for example, a lithium/air cell may have a discharge/charge efficiency of 50–60%, whereas a lithium intercalation cell typically has a discharge/charge efficiency of more than 80%). The energy efficiency of a Zn/air cell is particularly low because of its lower cell potential compared with that of the lithium/air cell. The air electrode in a metal/air cell is significantly different than that found at the positive electrode of a fuel cell because it needs to operate at potentials that can carry out both oxygen reduction and oxygen evolution. That is, it needs to be a bifunctional air electrode. This requirement eliminates as possible catalysts elements such as Pt and Pd that have been found to be effective for oxygen reduction because they form a surface oxide layer at the higher potentials at which oxygen is evolved and lose their catalytic activity (115, 116).

As with all practical batteries, more than just the active materials are required to make a practical cell. For example, a porous, electronically conducting framework is required for the air electrode. A catalyst can be deposited on or mixed with this framework to provide reaction sites with access to oxygen, a catalytic site, an electronically conducting network, and the ionically conducting electrolyte. The specific capacity of the positive electrode (in  $\text{mAh g}^{-1}$ ) is typically based on the weight of the carbon alone, which serves as a reaction site as well as an electronically conductive network.

A final important point that distinguishes metal/air cells from other types of cells is that they are not closed systems (unless an oxygen tank is carried). Therefore, both the gases needed for operation and possible contaminants may enter the electrodes and electrolyte.

In the remainder of this section we focus on the two most promising metal/air chemistries: the aqueous Zn/air system and the nonaqueous lithium/air system. Although an aqueous lithium/air



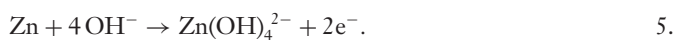
system can also be made, hydrogen evolution at the lithium electrode limits its reversibility. Because the potential of Zn is higher, hydrogen evolution is easier to suppress.

## Zinc/Air Cells

Zinc has been an important negative-electrode material in primary alkaline batteries for decades and has been paired with a variety of positive electrodes including nickel, iron, hydrogen, and air (117). Zinc/air cells are commercially available as the primary cells used in applications such as hearing aids. Practical cells have achieved specific energies greater than  $300 \text{ Wh kg}^{-1}$  (16). The major problem for the zinc/air cell is the difficulty of electrically recharging the system; indeed, the common alkaline ( $\text{Zn}/\text{MnO}_2$ ) cells can't be recharged for some of the same reasons. At the air electrode upon discharge, oxygen is reduced in the alkaline aqueous electrolyte,



The oxidation of hydroxyl ion to reform oxygen is possible, but the main problem in the cell is at the Zn electrode. There, upon discharge, Zn is oxidized to form zincate ion,



At the beginning of a discharge, zincate ion is soluble in the strongly alkaline aqueous electrolyte, but as the discharge proceeds and the concentration of zincate in the electrolyte rises, ZnO precipitates from the zincate throughout the cell,



Because the ZnO can precipitate wherever zincate concentrations significantly exceed the solubility limit, long-term cycling is limited, as the ZnO is likely to deposit in regions of the cell where it is disconnected from the electronic pathways of the electrode structure. The design of a reversible electrochemical system with a soluble product presents formidable challenges. A possible solution is to include additives that reduce the zinc solubility so that the product remains near the reaction site. Similar challenges are encountered in the Li/S system.

A second problem with the Zn electrode is that the Zn that is electrodeposited from zincate in solution can form dendrites between the negative and positive electrodes, possibly leading to a short circuit of the cell. Other challenges for the Zn/air system include the absorption of carbon dioxide by the electrolyte with subsequent crystallization of carbonate in the electrode structures and the drying out or flooding of the cell if it is left in a low-humidity or very high-humidity environment, respectively.

There are various ways to get around the problems of the Zn electrode and the bifunctional air electrode. For example, the cell can be mechanically recharged and the Zn regenerated externally. At the air electrode, separate electrodes can be used for the charge and discharge process, reducing the voltage window seen by individual catalyst materials. However, these schemes are unlikely to be suitable for use in an EV, and research into methods of reversibly cycling the Zn electrode (e.g., by designing an electrode or electrolyte that constrains zincate movement) and finding a stable and high-capacity bifunctional air electrode remain the best approaches.

## Lithium/Air Cells

Lithium is a highly reactive metal but, fortunately, forms protective films in both aqueous and nonaqueous systems. Li will react with water, but in the presence of  $\text{OH}^-$  ions at concentrations greater than 1.5 M it will form a protective film in a dynamic steady state. Although aqueous



lithium/air batteries exist, essentially no reversibility has been shown, and the electrolyte must be kept out of the system when not in use because of hydrogen generation at the Li metal electrode. Thus, here we consider only nonaqueous lithium/air designs. For example, lithium/air cells have been built with an electrolyte of polyacrylonitrile (PAN), ethylene carbonate, propylene carbonate, and  $\text{LiPF}_6$  (118). Other authors have also explored lithium/air cells (119–124). The reaction at the negative electrode upon discharge is the typical lithium oxidation reaction,



Although the reaction mechanism at the air electrode is still under investigation (124, 125), it appears that  $\text{Li}^+$  moves through the electrolyte and reacts at the positive electrode to form lithium peroxide,



Although concerns about Li metal forming dendrites during Li deposition are always present, the real challenge for the Li/air system is the air electrode. Unlike the Zn/air system, in which oxygen reacts to form  $\text{OH}^-$ , which then moves across the cell to the negative electrode, in the Li/air system the Li travels across the cell and is deposited in the positive electrode. Thus, whereas the Li electrode shrinks during discharge, the air electrode swells. The poor cycling seen in the Li/air cell is likely related to poor kinetics of  $\text{Li}_2\text{O}_2$  oxidation, formation of side reaction products [e.g.,  $\text{Li}_2\text{CO}_3$  (126)], and electronic isolation of  $\text{Li}_2\text{O}_2$ . One study found that whereas different catalysts had a minor effect on the voltage offset between charge and discharge, the catalyst did strongly affect the cyclability, indicating that the catalyst is important for the oxidation of  $\text{Li}_2\text{O}_2$  or other products back to  $\text{Li}^+$  and  $\text{O}_2$  (122). A significant offset between the charge and discharge curves occurs even at low current densities; an offset of 0.5 V is common. This limits the round-trip energy efficiency of the cell. The strong polarization is presumably related to the poor kinetics of oxygen reduction and  $\text{Li}_2\text{O}_2$  oxidation, although a true thermodynamic hysteresis may also be present.

A particular challenge for the design of a lithium/air cell is how to keep water out of the system when it is open to a humid environment; a membrane permeable to oxygen but impermeable to water may be necessary. Furthermore, although oxygen must be present at the air electrode, it should not be allowed to contact the surface of the Li electrode, as it would be quickly reduced. The presence of carbon dioxide at the Li electrode would also present problems (formation of  $\text{Li}_2\text{CO}_3$ ). In addition, the open system would need to deal with all other species that can be found in air, such as particulates,  $\text{NO}_x$ , and  $\text{SO}_x$ .

## Lithium/Sulfur Cells

The lithium/sulfur cell is an attractive candidate for development because sulfur is very inexpensive, produced in large quantities, and environmentally benign. The overall reaction upon discharge is:



This reaction has a very high theoretical specific energy of  $2600 \text{ Wh kg}^{-1}$ , compared with approximately  $600 \text{ Wh kg}^{-1}$  for the Li-ion cells now produced commercially. The lithium/sulfur cell was first explored as a high-temperature cell with a molten salt electrolyte in the late 1960s (127). Sulfur containment proved to be difficult, and long cycle life was not achieved. Subsequent work with  $\text{FeS}_2$  and  $\text{FeS}$  as cathode materials led to the development of Li/ $\text{FeS}_2$  cells for demonstration vehicles and for thermal batteries used in defense applications.

More recently several laboratories have investigated the Li/S cell using a variety of nonaqueous electrolytes (128, 129). One version of this system is under commercial development (130–132). Because sulfur has extremely low electronic conductivity, it is difficult to achieve high electrochemical utilization. During discharge of Li/S cells containing organic solvent-based electrolytes, lithium polysulfides form. These polysulfides are soluble in the organic solvents and can migrate away from the positive electrode, resulting in capacity loss and short cycle life. Several investigations into capacity loss rate reduction have been published. Some recent work on new families of electrolytes based on low molecular weight polymers and ionic liquids shows promise for achieving higher sulfur utilization and low polysulfide solubility (46, 133–135). Other approaches to longer cycle life include the containment of the sulfur in micropores of porous carbon (136).

If the development of Li/S cells is successful, there is the possibility to achieve approximately 800 Wh kg<sup>-1</sup>, which would be a large advance over existing battery technology in terms of cost, environmental impact, and performance. An all-electric vehicle with a range in excess of 300 miles per charge and a low price could be realized.

## CONCLUSIONS AND OUTLOOK

The development of batteries with high performance, long life, and low cost remains a challenge. Success in developing improved batteries will enable better electric and hybrid vehicles and reduce dependency on foreign sources of energy. The lithium-ion battery has opened the possibility of plug-in hybrid vehicles and all-battery vehicles. The next generation of high specific-energy cells requires that we move away from oxide- and phosphate-based systems toward lighter-weight reactants such as oxygen and sulfur. An important development in battery design is the increasing ability to carry out computations that aid in the material development process (28, 112). In some cases the computation of properties relevant to battery performance may be used to evaluate material suitability (137, 138).

Cost remains a key barrier. Developing higher specific-energy systems may allow a reduction in the weight of the battery, but going to a system with a higher specific energy, no matter what the size, offers challenges. In addition, it will be necessary to design and build a system around a chemistry that makes the battery completely safe.

## DISCLOSURE STATEMENT

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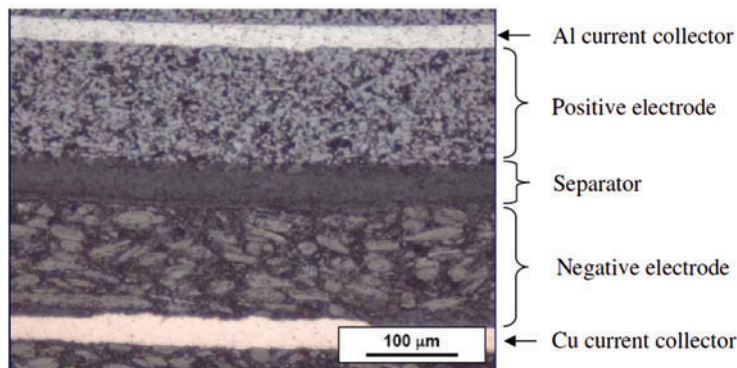
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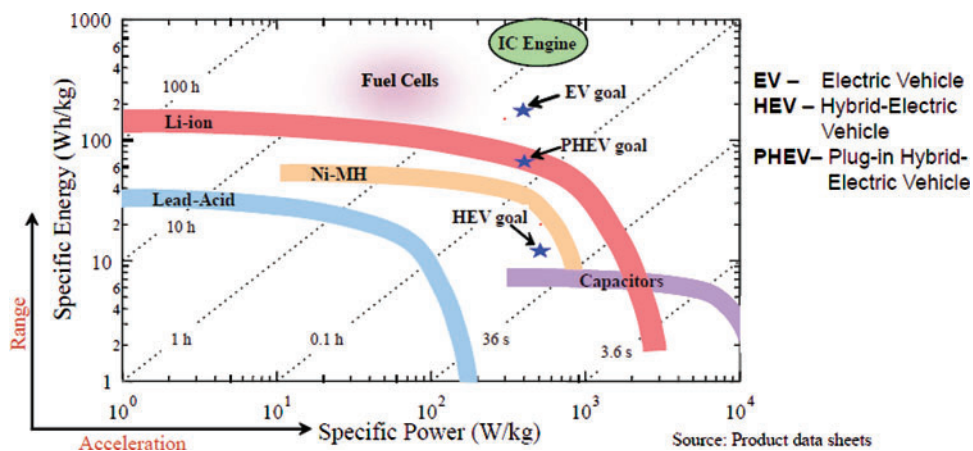
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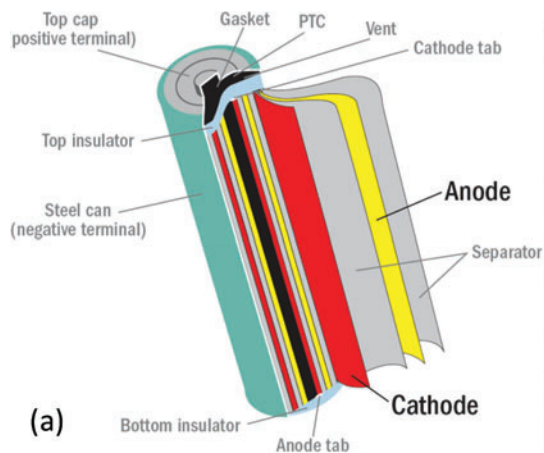
**Figure 2**

Cross-sectional image of a lithium-ion cell showing the principal components (two electrodes and a separator) of any cell. The image is from Quinn Horn of Exponent Consulting (21).



**Figure 5**

Ragone plot comparing the system-level performance of three battery chemistries, capacitors, fuel cells, and the internal combustion (IC) engine (13). Information is drawn from product data sheets. The Li-ion category is meant to reflect the performance of the  $\text{Li}_x\text{C}_6/\text{Li}_y\text{CoO}_2$  chemistry. Ni-MH: nickel-metal hydride.



**Figure 7**

(a) A spirally wound, cylindrical lithium-ion cell, 18 mm in diameter and 65 mm in length. (b) A Ni/MH prismatic module (containing six cells) from the Toyota Prius; it is 2 cm thick, 10.6 cm tall, and 28.5 cm long. The pressure release valve is in the upper right, and a thermal well is just to the left. Dimples on the surface improve heat transfer to air forced through the pack.



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## Errata

An online log of corrections to *Annual Review of Chemical and Biomolecular Engineering* articles may be found at <http://chembioeng.annualreviews.org/errata.shtml>