CAPACITOR ENERGY STORAGE FOR STATIONARY-POWER APPLICATIONS

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ABSTRACT

Electrochemical capacitors, sometimes called supercapacitors or ultracapacitors, have evolved through several generations of designs since the NEC Corporation, under license from SOHIO, first introduced their Supercap™ product in August 1978. This new energy storage device used highly-reversible charge storage in the electric double layer of a high-surface-area carbon, which provided unheard of capacitance density with essentially unlimited charge/discharge cycle life. Initially this technology was used to provide backup power for volatile clock chips and computer memories. Numerous other applications have emerged during the past 25 years including ones associated with portable wireless communication, power quality for distributed power generation systems, and high-efficiency energy storage for electric and hybrid gas-electric vehicles. This paper provides basic information for stationary-power technologists interested in exploiting electrochemical capacitor technology.

INTRODUCTION

Electrochemical capacitors (ECs), also referred to as supercapacitors or ultracapacitors, store electrical energy as charge in a highly reversible manner. They were invented by Standard Oil of OHIO[1] in the 1960s and under license commercialized in Japan for computer memory backup applications. More recently several companies, mostly outside the US, have developed larger ECs that are suitable for various transportation and power quality applications.

ECs have been recognized for several years as having superior performance to competing battery technology in some automotive applications[2,3]. Capacitors have exceptional power performance, high cycle life, long operational life, require no maintenance, and can be safer and more environmentally friendly than batteries. Large ECs have been successfully used in many demonstration projects, but their cost has been too high for widespread acceptance. Their use was first proposed in the early 1990's as a load-leveling technology for electric vehicles (EV's)[4]. They can offer performance advantages over batteries in load-leveling by providing higher power for acceleration, larger energy capture efficiency during braking, higher cycle life, and longer calendar life.

These advantages of a capacitor in an EV generally apply to hybrid electric vehicles now on the market (with batteries), such as the Toyota Prius and Honda Insight and Civic. For example, the Prius batteries presently recaptures < 50% of the braking energy whereas capacitors could recapture > 90% of the braking energy. Another application is in "minimal hybrid" vehicles where there is no electric drive but the engine is stopped and restarted when the vehicle stops, for example at a stoplight. Fuel cell powered vehicles, and fuel cells in general, typically display slow response to power level changes and thus are oversized to provide responsive acceleration. The fuel cell size could be reduced when paired with an electrochemical capacitor to optimize energy and power requirements for the entire system. Honda has built and is presently evaluating a fuel cell–electrochemical capacitor power system in demonstration vehicles. Oshkosh Truck is developing a heavy-vehicle hybrid power system for use in refuse trucks, where 800-1000 start-stop cycles occur each day. The military is interested in replacing its fleet of five and seven-ton trucks with hybrids for increased gas mileage. In each of these cases an advanced electrochemical capacitor offers significant performance and life advantages over a battery.

Capacitors have also been evaluated for supplying the power for a number of advanced vehicle applications not related to the drive train. These applications include electric power steering, preheating of the catalytic converter, active suspension systems, electrically operated air conditioners, and electromagnetic valve actuation, all of which are being considered to improve vehicle gas mileage, reduce emissions, and/or improve performance. Even more applications exist in a 42 V automotive electrical system that is expected to replace the present 12 V systems in the near future. A cost competitive EC is highly desired to supply the power necessary to operate these additional electric loads.

Electrochemical capacitors are already being test marketed to the trucking industry for diesel engine starting[5-9]. Increased use of ancillary electrical equipment, such as computer systems and driver conveniences, can drain the truck batteries during extended key-off stops so they fail to start the truck, especially when cold. An EC improves the reliability and life of the starting system. This market is expected to increase as no-idle laws are enacted. ECs would gain widespread acceptance for this application if their price were more competitive with the cost of lead acid batteries.

A similar but non-vehicle application is engine starting of stationary motor generator sets like those used in many public buildings during loss of grid power. The starting batteries currently used are usually large NiCd or lead acid systems. A capacitor system can be more reliable, requires no maintenance, and is environmentally friendlier than either of these battery systems. Capacitors also recharge more quickly than batteries and require no special precautions to vent flammable gases during charging as is often mandated for batteries.

Electrochemical capacitors have been used successfully in demonstration projects to form large, high-voltage energy storage systems. Example systems include a 1.5 MJ, 400 V gas-electric hybrid bus;[10] 30 MJ, 190 V all-electric trucks and buses;[11] 500 kJ, 650 V backup adjustable speed drive;[12] and a 1 MJ, 100 kW uninterruptible power supply (UPS) system.[13] ECs are being considered by NASA for a very large, ~270 V power source for the next-generation reusable launch vehicle;[14] and for a 313 kVA, 480 V distributed-generation power load leveling power system.[15] ECs have demonstrated certain advantages over alternative approaches in most of these applications.

Considerable effort is presently underway by most manufacturers to make ECs more cost competitive with the alternative solutions. Should low-cost ECs be developed and reach large-volume production, then this may create a path for EC use in stationary bulk energy storage systems. That market size would vastly exceed the transportation market size. This paper aims to help prepare the stationary-power industry for the coming of ECs. Topics that follow include capacitor basics, electrochemical capacitor technology development, product designs, health and safety issues, comparison with battery technology, and finally a list of large EC producers with contact information.

CAPACITOR BASICS

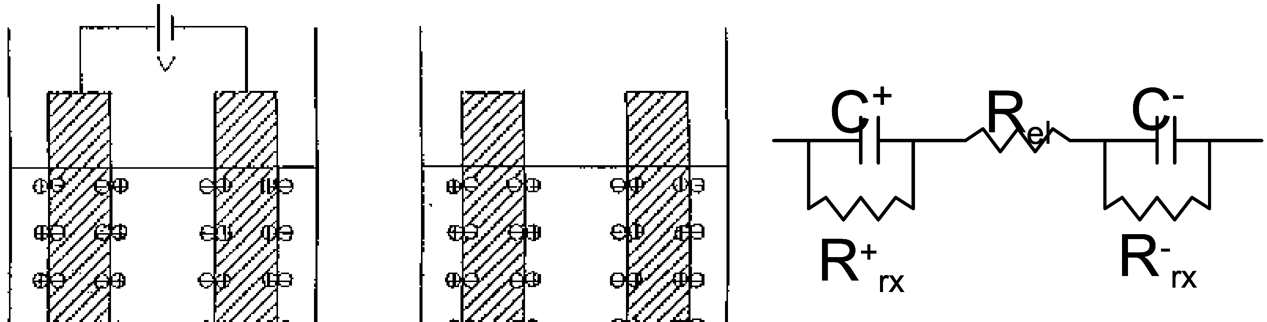
A capacitor is a device used for storing electrical charge. Generally the charge is stored physically, as equal quantities of positive and negative charge separated to opposite faces of an insulating material. When the two faces are connected by an external current path, current flows and work can be performed until complete charge balance is achieved. The capacitor can be returned to its charged state, for instance, using an applied voltage. Discharge and charge can be repeated over and over again since the process is highly reversible, i.e. charge is stored physically--no chemical or phase changes occur in the device. The quantity of charge **Q** stored in a capacitor is equal to device voltage **V** times a proportionality constant **C** called capacitance $\mathbf{Q = C\ V}$. An uncharged capacitor has no voltage. The charge **Q** can be related to the current **i** by the equation $\mathbf{Q = i\ t}$ where **t** is time. Then with constant-current charging $\mathbf{i_o}$ the capacitor voltage increases in time **t** in a linear manner $\mathbf{V = (i_o/C)\ t}$. Similarly, voltage declines in a linear manner during constant-current discharge. Capacitor voltage cannot increase without limit during charging. At some voltage a "breakdown" occurs within the insulating material that stops the charging process. This breakdown is sometimes observed as a spark in the material. Some capacitor types are limited to ~1 V per unit cell while others have much higher voltage limits, for instance more than 10,000 V.

The fundamental equation relating the capacitance of a capacitor to its construction is $\mathbf{C = k\ \varepsilon A/d}$ where **k** is a constant, ε is the dielectric constant of the insulator, **A** is its area, and **d** is its thickness. Thus high capacitance is achieved by using an extremely thin, large-area insulating material having a high dielectric constant.

Because of the physical charge storage, a capacitor's charge and discharge rates can be extremely high, allowing some "electrostatic type" polymer film or ceramic capacitors to operate at gigahertz frequencies (~10^{-9} seconds charge time). This type capacitor has the lowest energy density of the three types. "Electrolytic type" capacitors can have more than ten-times higher energy density, which is achieved through the use of a thinner insulator, grown starting at zero-thickness on a rough metal surface like etched aluminum. These capacitors generally can be used at frequencies below 10 to 50 kHz.

"Electrochemical type" capacitors are the third type, having the distinction of providing the highest specific energy, often 100- to 1000-times greater than electrolytic capacitors[16]. This is achieved first by exploiting the naturally-formed "double layer" insulator at a solid-liquid interface when voltage is applied and second, use of high-surface-area electrode material like activated carbon. The thickness of the double layer is approximately one nanometer and the surface area of activated carbon is ~1000 m^2/g of material, creating a device with specific capacitance values of ~50 F/g or more!

Referring to **Figure 1**, a simple EC can be constructed by inserting two conductors in a beaker containing an electrolyte, say carbon rods in salt water. Initially there is no measurable voltage between the two rods. But when voltage is applied between the rods, charge separation occurs at each liquid-solid interface, effectively creating two capacitors that are series-connected by the electrolyte and shown by the equivalent circuit in Figure 1. The charge separation persists after removal of the voltage source--a capacitor charge storage device has been created. Practical ECs use high surface area activated carbon with a highly conductive electrolyte. Carbons typically have specific capacitance values of 50 to 100 F/g, creating the possibility for capacitor cells rated at many thousands of farads.



$$\frac{1}{C_t} = \frac{1}{C_+} + \frac{1}{C_-}$$

Figure 1: Demonstration of double layer charge storage at the interface of each electrode in an electrolyte after voltage is applied (left). When removed (right), the charge separation persists on each electrode. Two capacitors in series with the electrolyte are created. The equivalent circuit model is shown on the right, with the equation used to calculate the total device capacitance C_t. Practical ECs use high surface area carbons for charge storage rather than planar surfaces.

With this capacitor type that has exceedingly high capacitance density comes some less desirable features. One is low operating voltage, typically 1 to 3 V per cell. This necessitates using strings of series-connected cells to meet the voltage requirements of many practical applications[17-18]. A second less desirable feature is poor frequency response, with operation generally limited to 100 Hz or less, and more often <1 Hz. Compared with other capacitors types, electrochemical capacitors are effectively dc devices.

It is informative to compare the size, mass, cost, and characteristic time response of energy storage systems constructed with the different capacitor types. These values were calculated using the largest size commercial examples identified. This information is summarized in **Table I** for systems that store 1 MJ (0.3 kWh) of energy. Electrochemical capacitors are at least ten-fold lower in mass, volume, and cost than the other capacitor types. They are vastly "slower" in response than other types. This slow response precludes their use in ac filtering applications. Thus, electrochemical capacitors generally do not compete for applications usually served by electrolytic capacitors. Nevertheless, electrochemical capacitors have extremely fast response compared with a battery—3 second response time corresponds to a 1200C rate using battery terms.

Table I: Typical characteristics of capacitor types for a system that stores 1 MJ (0.3 kWh) of energy. The electrochemical capacitor type has extraordinary energy density compared with the other types, but very poor time response. This last factor precludes their use in conventional ac filtering applications.

Capacitor Type	Mass (kg)	Volume (m^3)	Cost (k$)	Time response (s)
Electrostatic	200,000	140	700	10^{-9}
Electrolytic	10,000	2.2	300	10^{-4}
Electrochemical	30 to 100	0.02 to 0.1	2 to 20	0.3 to 3

Table I shows distinguishing features of the three types of capacitors. Some specialized products fall outside the ranges listed, for instance a pulse electrochemical capacitor with 10 ms response time. Such enhancements, however, reduce the specific energy significantly.

ELECTROCHEMICAL CAPACITOR TECHNOLOGY DEVELOPMENT

Electrochemical capacitors have evolved through several generations of designs during the past 25 years.[19] Symmetric designs, where both positive and negative electrodes are made of the same material with approximately the same mass, were the earliest type available, first with aqueous electrolyte then, to achieve higher cell voltage, with an organic electrolyte. Asymmetric designs have different material for the two electrodes, with one of the electrodes having much higher capacity than the other. These types offer certain performance advantages and are now becoming available.

Initially EC technology was used to provide backup power for volatile clock chips and computer memories. Numerous other applications have emerged including ones associated with power pulses for portable wireless communication, reliable cold-weather diesel engine starting, power quality for distributed power generation systems, and high-efficiency energy storage for electric and hybrid gas-electric vehicles. Capacitors for power quality and transportation are physically very large compared with the first products and often can store many megajoules rather than joules of energy.

The first generation capacitors used a symmetric design with activated carbon for the positive and negative electrodes, each with approximately the same mass and similar capacitance values. The choice of electrolyte was an aqueous solution, usually composed of sulfuric acid or potassium hydroxide. Because of the aqueous electrolyte, operating voltages were limited to ~1.2 V per cell, with nominal ratings of ~0.9 V.

The second generation EC was similar to the first, but with an organic rather than an aqueous electrolyte. The organic electrolyte typically is an ammonium salt dissolved in an organic solvent such as propylene carbonate or acetonitrile, which allows operation at higher unit cell voltages. This is the most popular type today, with rated voltages in the range of 2.3 to 2.7 V/cell, depending on manufacturer.

Operation at higher voltages offers distinct energy and power density advantages along with some disadvantages. The dielectric constant of the organic solvent is less than that of water; the double layer thickness is greater because of the larger-size solvent molecules; the effective surface area of the electrode is somewhat diminished because the larger ion sizes cannot penetrate all pores in the electrodes; and the ionic conductivity of the electrolyte is much less than that of aqueous electrolytes, particularly at low temperatures. Stable, long term operation at higher voltages requires extremely pure materials: trace quantities of water in the electrolyte, for instance, can create problems. Thus, the device must be packaged in such a way that water does not enter the capacitor. The net effect of using an organic rather than an aqueous electrolyte is increased energy density.

The most recent type of EC is referred to as an asymmetric capacitor.[11,20] They are comprised of two capacitors in series, one being electrostatic and the other faradaic. The electrostatic capacitor is exactly like those used in the symmetric devices. It consists of a high-surface-area electrode with double layer charge storage. The faradaic electrode relies on an electron charge transfer reaction at the electrode-electrolyte interface to store energy, exactly like an electrode in a rechargeable battery. In this design the capacity of the battery electrode is typically many times greater than the capacity of the carbon electrode. Thus, the depth of discharge of the battery electrode is very small during operation, allowing higher cycle life. Different asymmetric capacity ratios are used depending on the specific application. Asymmetric ECs having aqueous electrolytes have an important advantage of voltage self-balancing, as do some aqueous secondary batteries. This is achieved by operating on a gas cycle to prevent overcharge. Thus, a series-string of cells can be equalized by applying a slight overvoltage.

Several asymmetric design capacitors using an organic electrolyte are under development. This design provides an opportunity for the higher operating voltage afforded by an organic electrolyte. One example of this design is an activated carbon electrode mated with a lithium ion battery intercalation electrode.[21] Energy densities >14 Wh/kg have been reported in such prototype devices. A magnetite battery-like electrode in combination with activated carbon is a second example.[22] Research on this type of electrochemical capacitor using low-cost materials is very active at present.[23]

ELECTROCHEMICAL CAPACITOR DESIGNS

The carbon electrodes used in both symmetric and asymmetric electrochemical capacitors consist of a high-surface-area activated carbon having area on the order of 1000 m^2/g or more in particulate, plastic bonded, or cloth form. The carbon electrode is in contact with a current collector. A separator material is used to prevents physical contact (shorts) between the two electrodes, but allows ion conduction. One design utilizes particulate carbon pasted on aluminum foil in a spiral-wound configuration. Such construction can be performed on a high-speed winding machine. While this construction lends itself to a right-cylinder product, it can also form rectangular packaging, a form factor more desirable for some applications. Present asymmetric electrochemical capacitor cells using aqueous electrolyte are constructed in a similar manner to aircraft NiCd battery products. The first commercial product of this type used a nickel-oxyhydroxide positive battery electrode, an activated carbon cloth negative electrode, and aqueous potassium hydroxide electrolyte.[11]

Figure 2 shows photographs of representative capacitors from the eight companies that manufactures large-size products. The ELIT and ECOND capacitors have a symmetric design, use an aqueous electrolyte, and have bipolar construction. Thus, these devices have many series-connected cells within their package. The ESMA capacitor is a module of asymmetric cells, but interconnected externally. The Okamura module is composed of many individual symmetric cells externally connected. This module also contains cell voltage-balance electronics. And last are the individual cells made by NESS, EPCOS, Panasonic, and Maxwell. They use symmetric design with a wound construction.

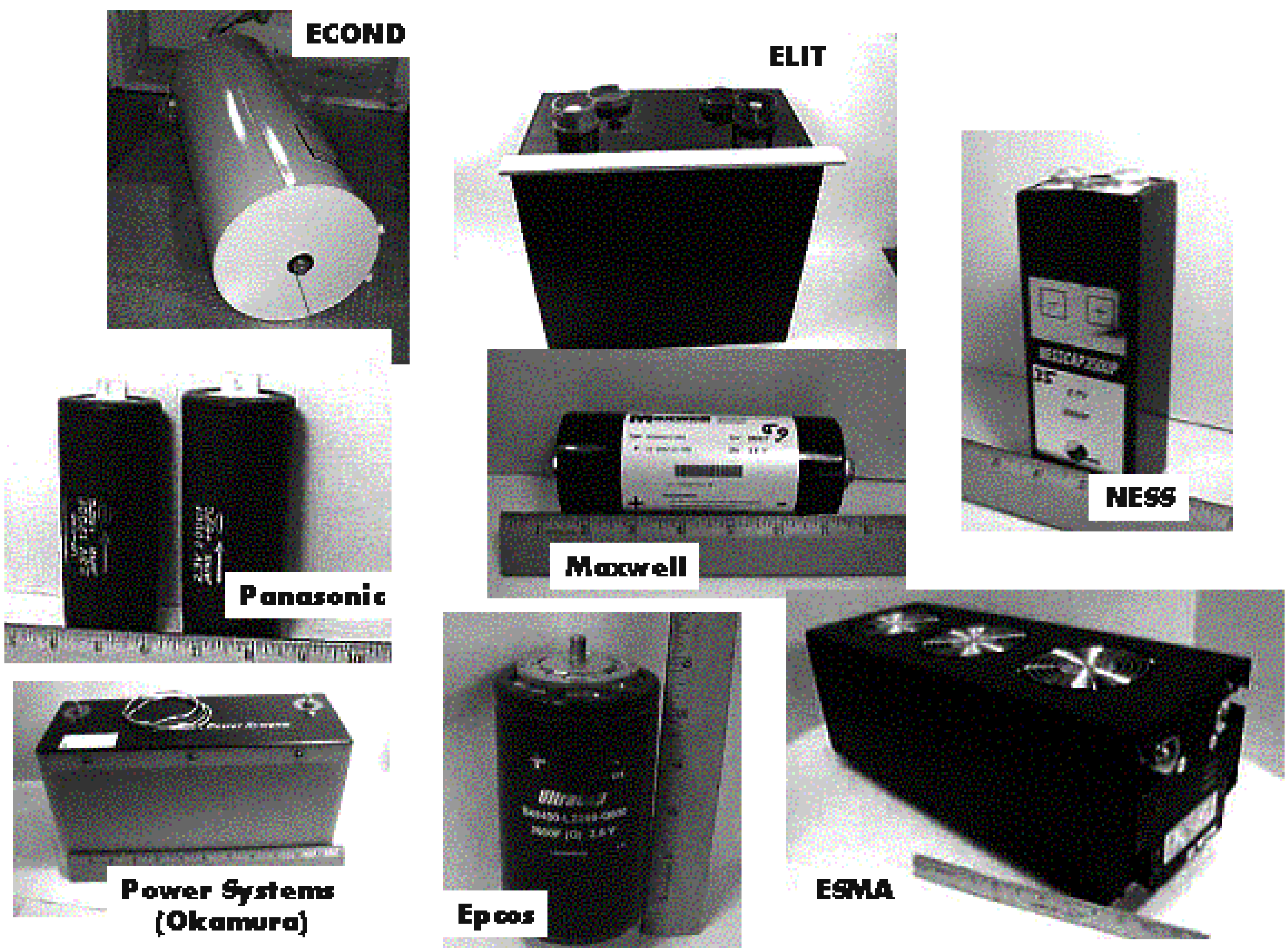


Figure 2: Commercial capacitor examples from the eight companies making large ECs. Notice the 12” ruler in each photo.

The electrolyte of an electrochemical capacitor is an important constituent. Properties most desired include high conductivity and high voltage stability. Little can be done to change the conductivity and voltage characteristics of aqueous-based electrolytes, but major improvements appear possible for the symmetric capacitors having non-aqueous electrolyte. Higher-conductivity electrolyte yields increased power performance, and high voltage stability allows stable operation at high voltage. Popular electrolytes are comprised of an ammonium salt with a solvent like propylene carbonate, dimethyl-carbonate, or acetonitrile. At the present time, acetonitrile is the most popular solvent in large capacitors. It offers higher operating power, but at the expense of using a toxic and flammable material.[24]

One feature common to all electrochemical capacitors is the requirement that some pressure be applied to the cell so that its electrodes remain in contact with the separator, that the electrodes are in contact with the current collectors, and that everything is wetted with electrolyte. The amount of pressure required depends on the design and electrode form. Winding pressure is typically adequate for cylindrical devices. External pressure plates are often used for prismatic devices.

HEALTH, SAFETY, AND ENVIRONMENTAL ISSUES

Capacitor safety issues can be grouped into electrical, chemical, and fire and explosion hazards. Electrical hazards are very similar to those of batteries. Hazards from chemical burns and chemical exposures can be similar to some batteries. For instance, ECOND, ELIT, and ESMA electrochemical capacitors use potassium hydroxide electrolyte, which is the same electrolyte used in alkali primary cells and nickel-metal hydride secondary batteries. An unknown safety-related issue arises because acetonitrile, a.k.a. methyl cyanide, is the electrolyte solvent in some large symmetric, organic electrolyte capacitors. Use of this chemical has not been fully evaluated for potential problems in large-scale applications.

Whenever there is a concentrated quantity of stored energy, the possibility always exists of creating high temperatures that can lead to combustion. Fire hazards are minimal for aqueous electrolyte products, and similar to batteries for some organic electrolyte products.

Maxwell, Panasonic, NESS, EPCOS, and ECaSS capacitors contain carbon, aluminum, a paper or polymer separator, rubber, and an organic electrolyte. Only the aluminum appears to be suitable for recycling. ECOND, ELIT, and ESMA capacitors contain carbon, nickel, plastic, potassium hydroxide electrolyte, and steel packaging. Nickel, plastic, and steel appear suitable for recycling. These products would probably qualify to enter the nickel-based battery waste stream.

ELECTROCHEMICAL CAPACITOR COMPARISON TO BATTERIES

Batteries and capacitors have significant differences.[25] A fundamental difference between the two is that the battery stores energy in chemical reactants capable of generating charge, while a capacitor stores energy directly as charge. This difference is significant in that battery discharge rate, thus power performance, can be limited by reaction kinetics of the battery electrodes and mass transport of reactants. This often manifests itself as a charge rate that is much lower than the discharge rate. Further, thermodynamics of the battery reactants dictate system operating voltage, which is relatively constant and thus to first order independent of the state of charge (SOC) of the battery. Consequently, it is often difficult to precisely measure the SOC of a battery. And finally, chemical reactions in a battery are not fully reversible, meaning charge/discharge cycle life is limited in number. This number is one cycle in a primary battery! It can be many hundreds of cycles in secondary batteries, depending on chemistry and design.

In contrast, reaction kinetics are not involved with symmetric ECs so they do not limit charge and discharge rates, creating the possibility for exceptionally high power capability for discharge <u>and</u> charge. This is essentially true for asymmetric designs also because the faradaic electrode always has a very shallow depth of discharge due to the capacity asymmetry. And as discussed above, capacitor voltage is not constant but rather starts at 0.0 V for zero SOC and increases linearly with charge to its rated voltage. Thus, capacitor SOC is always known. But this sloping discharge often means additional complexity and cost in the load electronics. And finally, capacitors store charge physically rather than chemically and therefore experience no volume or phase changes. Then charge storage is highly reversible, providing a capacitor with essentially unlimited charge/discharge cycle life.

A major specific property difference between a battery and a capacitor is energy density. Chemical reactions in the battery mean high energy storage density. Physical charge storage in the capacitor means lower energy density. A basic reason for this difference is that most of the material in a battery electrode can contribute to storage while just the surface contributes to storage in a capacitor. Battery specific energy values range from ~25 Wh/kg to more than 100 Wh/kg while large electrochemical capacitors have specific energy values in the 1 to 10 Wh/kg range, depending on design and optimization.[26]

Batteries and electrochemical capacitors both have relative low-voltage cells, requiring use of series strings of cells to meet the requirements of high-voltage applications.

In summary, batteries have high specific energy, flat discharge curves, charge/discharge rate limitations, cycle-life limitations, and require maintenance. Capacitors have lower specific energy, a sloping discharge curve that directly provides SOC, essentially unlimited cycle life, high power performance, and maintenance-free operation.

LARGE ELECTROCHEMICAL CAPACITOR PRODUCERS

Commercial manufactures of large electrochemical capacitors are listed in **Table II**, with country of origin, size of a typical product, and website. Each of these companies has brochures and product information. They all provide application engineering services to assist users with product selection and information on the best mode of use for optimum results.

Table II: Companies that manufacturer large electrochemical capacitors. The first two listed rely on bipolar construction for their products, and the others construct single cells that can be connected in series or parallel.

Manufacturer	Country	Typical Product Energy and Voltage Ratings	Website
ECOND	Russia	40 kJ, 14--200 V	www.tavrima.com
ELIT Stock Company	Russia	50 kJ, 14--200 V	www.elit-cap.com
EPCOS AG	Germany	15 kJ, 2.5 V 40 kJ, 14 V	www.epcos.com
ESMA Joint Stock Company	Russia	20 kJ–1.2 MJ, 14 V 30 MJ, 180 V	www.esma-cap.com
Maxwell Technologies, Inc.	USA, Switzerland	8 kJ, 2.5 V	www.maxwell.com
NESS Capacitor Company	Korea	18 kJ, 2.7 V	www.nesscap.com
Okamura Laboratory, Inc. with license to others	Japan	5 kJ, 2.7 V 100 kJ, 54 V	www.okamura-lab.com
Panasonic	Japan	6 kJ, 2.5 V	www.maco.panasonic.co.jp

SUMMARY

Electrochemical capacitor technology has advanced considerably in recent years, making kJ devices common today. Their exceptional performance has been clearly demonstrated in many applications. The hope is to find a large-volume transportation application that will allow significant cost reductions so as to make the ECs more competitive in other applications. Then ECs may be in a position to start capturing some part of the stationary-power market.

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