

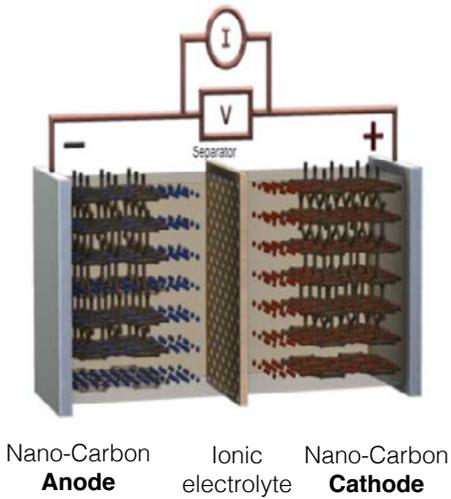
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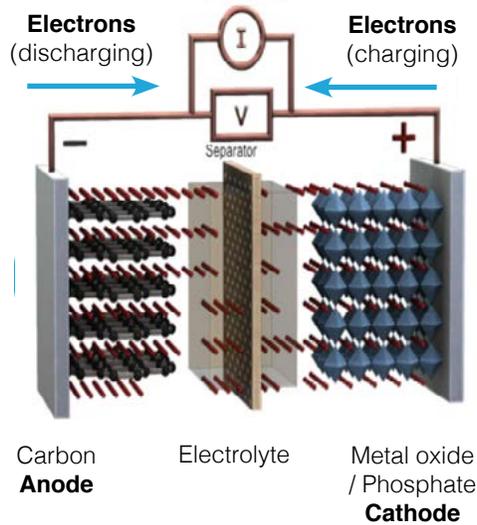
Carbon-Ion™:

A NEW, SAFER & FASTER CHARGING CATEGORY OF RECHARGEABLE ENERGY STORAGE DEVICES

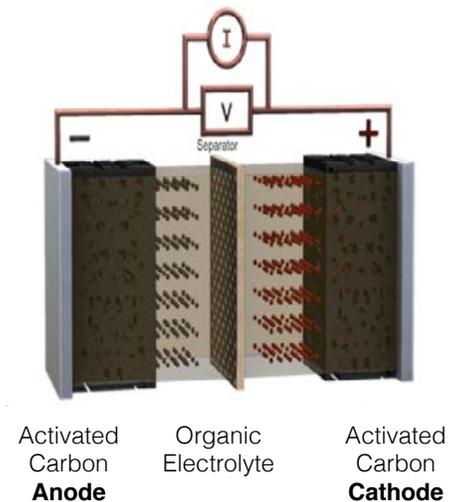
## CARBON-ION CELL



## LI-ION BATTERY



## SUPERCAPACITOR



# Carbon-Ion: a new, safer and faster charging category of rechargeable energy storage devices



## ABSTRACT

Lithium-ion (Li-ion) batteries are widely used in mobile phones, laptop computers, cordless appliances and electric vehicles because they have a high energy density. However, they contain a highly flammable electrolyte and in addition to depositing lithium, cathodes can become thermally and structurally unsafe if overcharged. This resulted in the recent high profile Samsung Galaxy Note 7 product recall. [6]

Much work is going on to improve the energy density of lithium-ion batteries using different anode and cathode chemistries. These include Si-based anodes, nickel rich cathodes (e.g. NCA, NMC 811) and different cell fabrication formats, such as solid state batteries with lithium metal anodes. To combat the rate issue, lithium titanate and LFP have been used. But for every drawback that is addressed, there is another downside, for example higher costs, lower voltage, temperature of operation, shorter lifetime and reduced safety.

Supercapacitors, also known as ultra-capacitors or electrochemical capacitors or electrical double layer capacitors (EDLCs) are used for storing and delivering electrical energy orders of magnitude faster than batteries. They also have a very long life, offering 100,000+ charge/discharge cycles compared

to Li-ion at typically just 1,000. However, supercapacitors have low energy densities so have not been used to store energy over a long period of time.

Commercially available supercapacitors are constructed using activated carbon with an organic electrolyte. They do not contain lithium, but are still flammable because of the solvents mixed with the electrolytes.

There is a new category of energy storage that uses nano-carbons and ionic electrolytes. This allows the device to work safely at higher voltages, which in turn offers the potential to increase energy density and bring it closer to that of a lithium-ion battery. These nano-carbon systems could also meet or exceed the energy density of other rechargeable batteries such as nickel cadmium and nickel metal hydride. This has the added advantage of retaining the rapid charging and long life of commercially available supercapacitors, and with no fire risk.

This paper presents a status report on C-Ion and the importance of incorporating advanced nano-carbons, new electrolytes and improved fabrication techniques for improved energy density.



## INTRODUCTION

**Electrical energy storage devices such as lithium-ion (Li-ion) batteries, supercapacitors, and carbon-Ion (C-Ion) differ from each other in terms of the amount of energy that can be stored (energy density), the time it would take for storing and delivering energy, and the usable life of the devices. [1-5]**

### RELATIVE PERFORMANCE CRITERIA OF ENERGY STORAGE DEVICES

	LITHIUM-ION	SUPERCAPACITOR (EDLC)	CARBON-ION
Energy Density	High	Low	Medium
Charging	Slow	Fast	Fast
Discharge rate	Slow	Fast	Fast
Cycle life	Low	High	High
Safety	Flammable	Flammable	Non-flammable
Recycle	Poor	Fair	Good



## MARKET DEMAND

Li-ion batteries are used extensively in mobile phones, laptop computers, cordless power tools, cordless cleaners and electric vehicles; a market expected to be worth \$140 billion by 2026 [5]. Used correctly they are perfectly safe, however if charged incorrectly Li-ion batteries can go into thermal runaway. In October 2016 Samsung [6] had to recall their Galaxy Note 7 mobile phone, at a cost of billions of dollars. The cause of this failure has been attributed to a design in which the battery was too thin and packed in too tightly to allow for pressure or expansion. [6]

Much work is going on to improve the performance of lithium batteries using different chemistries such as lithium titanate that improve the rate of charge, and solid state lithium batteries that can improve energy density. But for every drawback that is addressed, there is another downside. For example, higher cost, lower voltage, temperature of operation, shorter lifetime of use or the fact the electrodes in the batteries expand and contract during charging and discharging.

Supercapacitors, also known as ultra-capacitors or electrochemical capacitors or electrical double layer capacitors (EDLCs) are used for storing and delivering electrical energy orders of magnitude faster than batteries. They also have a very long life, offering 100,000+ charge/discharge cycles compared to Li-ion at ~1,000. However, supercapacitors have low energy densities so have not been used to store energy over a long period of time.

Today, there are commercially available supercapacitors that do not contain lithium. Most of these supercapacitors are constructed by winding a pair of carbon coated aluminum foils with a polymeric separator sandwiched in between, and by filling the cell with an organic electrolyte. Activated carbon is usually used as the electrode material, and the operating voltage of the cell is typically 2.7V (Volts). Despite high power capabilities, these supercapacitors have less than 5% of the energy density of a Li-ion battery [4].

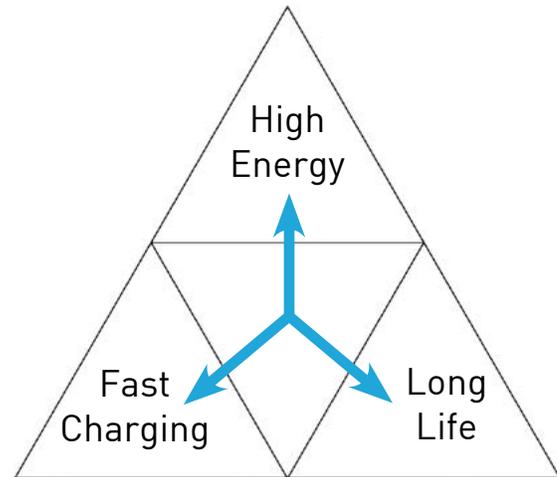
Engineers are currently working on nano-carbons and ionic electrolytes which permit a higher voltage system. This provides higher energy densities than conventional supercapacitors, with charging rates and cycle life that outstrip Li-ion batteries, in a form that can be safely transported around the globe.

This means C-Ion cells can be used either on their own or in combination with Li-ion batteries for a range applications such as (a) cordless power tools or appliances that charge in minutes, (b) mobile phones or laptop computers where the charge time could be reduced from hours to minutes, (c) infrastructure energy storage such as in building emergency lights or solar panels (where the long life time of C-Ion means there is no requirement to replace them every few years) and (d) to store energy rapidly in hybrid and electric vehicles.



## THE ENERGY STORAGE 'TRILEMMA'

Designers of systems that require rechargeable batteries have long faced the energy storage trilemma described by IDTechEx [1] in Figure 1, where it has been possible to achieve any two of these goals but not all three.



**Energy storage trilemma** source: IDTechEx [1]

Ragone [2-3] describes lithium-ion (Li-ion) batteries with a high energy density up to 200Wh/kg (Watt hours per kilogram) and with low power density of less than 1 kW/kg (Kilo-Watt hour per kilogram). Today, most commercially available supercapacitors can deliver orders of magnitude higher power densities (10 kW/kg and above), but with lower energy densities of around 5Wh/kg when compared to Li-ion batteries [4].

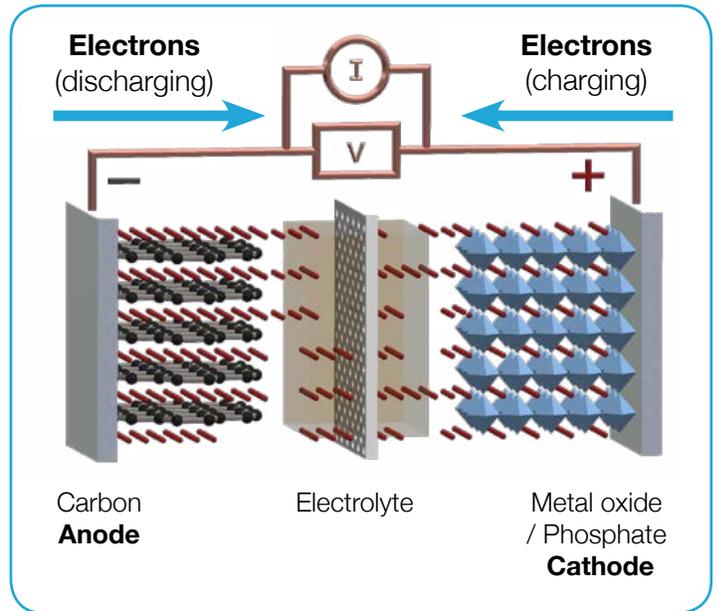


# LI-ON BATTERIES

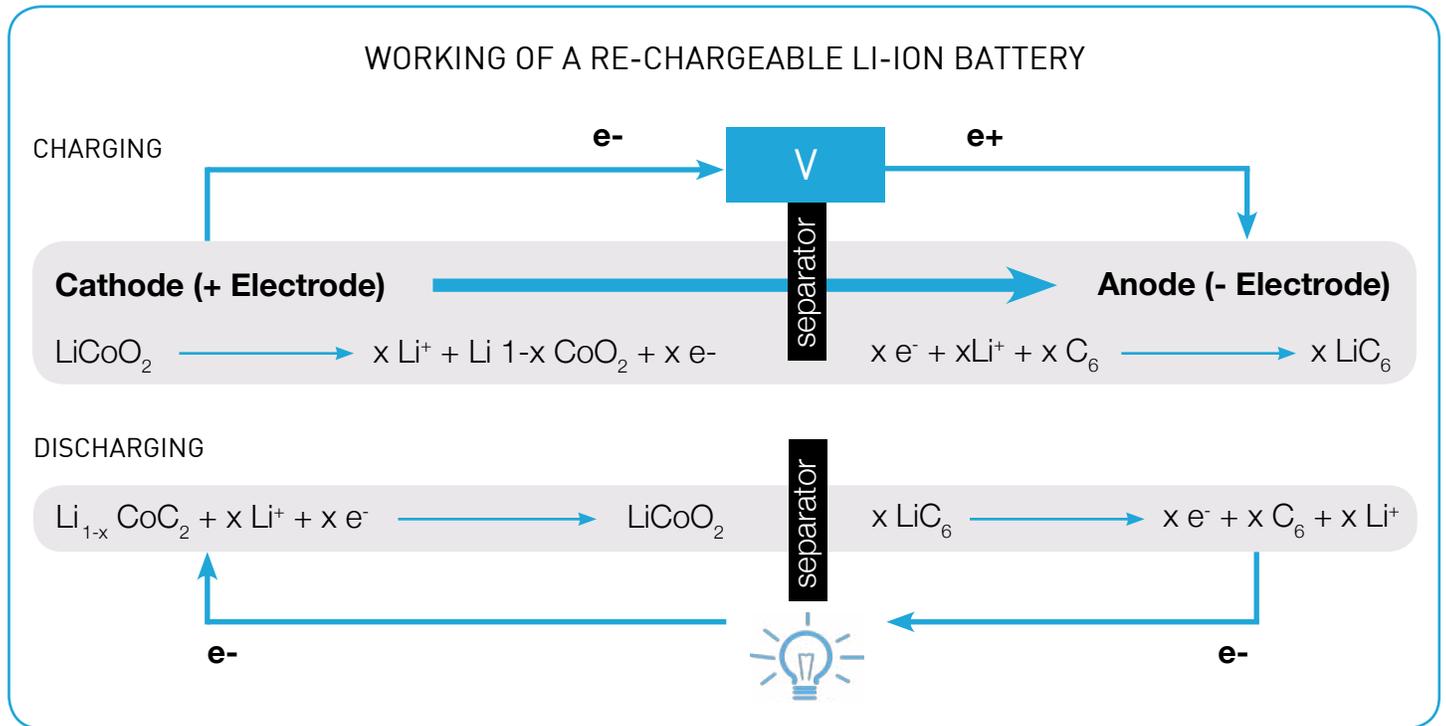
Li-ion batteries usually contain lithium containing oxide or phosphate working as a cathode (+ electrode). The commonly used cathode materials are  $\text{LiCoO}_2$ ,  $\text{LiNi}_{1-y-z}\text{Mn}_y\text{Co}_z\text{O}_2$ ,  $\text{LiFePO}_4$  and  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ . Graphite is the commonly used anode (- electrode). Organic solvents containing lithium compounds are used as electrolytes. During charging, lithium ions are de-intercalated (removed) from the cathode and released into the electrolyte. These lithium ions migrate to the anode and intercalated (inserted) into the graphite anode. Figure on the right is a schematic of a Li-ion cell.

The removal of lithium from the cathode induces a proportional amount of, for example,  $\text{Co}^{4+}$  in the resultant  $\text{Li}_{1-x}\text{CoO}_2$  to maintain charge neutrality.

When the battery is discharged, the lithium ions are released (de-intercalated) from the graphite anode and shuttle back to the cathode, where they are inserted (intercalated) into the crystal lattice. The de-intercalation of lithium ions from the graphite anode is accompanied by concurrent release of electrons and these electrons flow through the external lead.



The figure below portrays the electrode reactions that occur during charging and discharging. It illustrates the reactions at the cathode and anode, and depicts the 'shuttling' of lithium ions back and forth during the charging and discharging process.



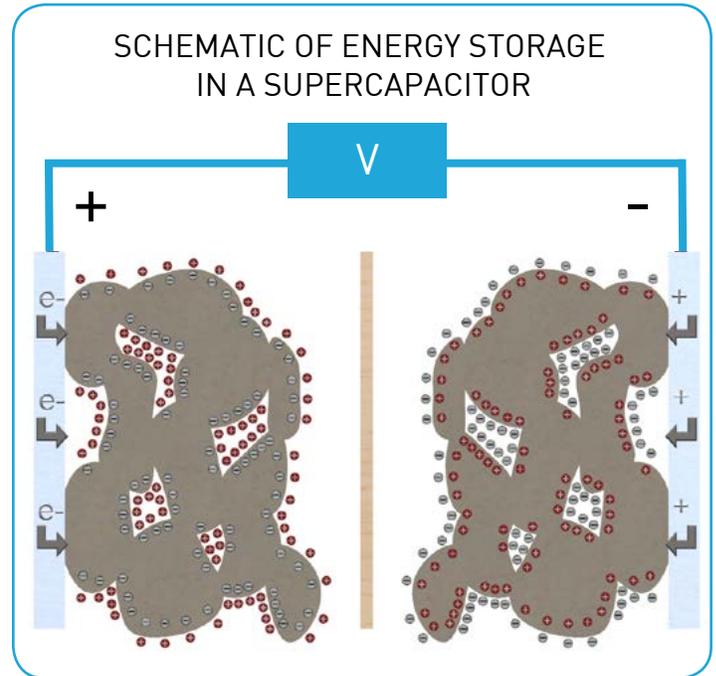
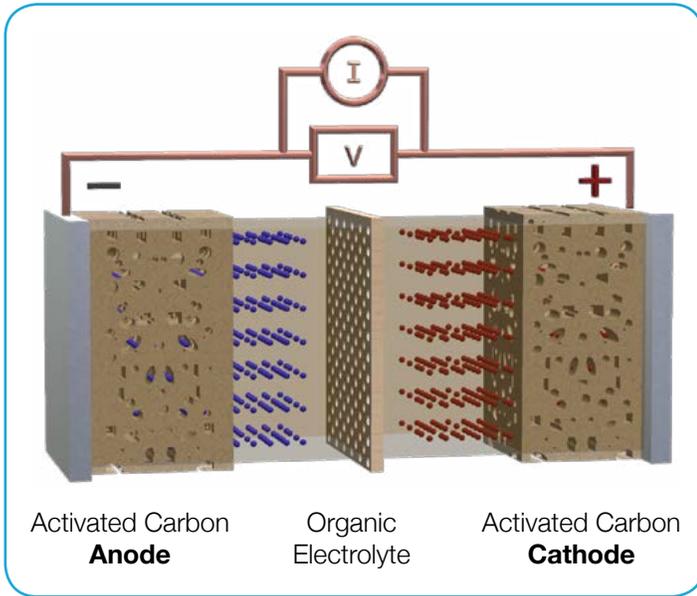
The cathode and anode materials are capable of accommodating and releasing lithium ions. The layer structure of both cathode and anode materials are flexible to reversibly intercalate lithium ions in the inter-layer space, without undergoing any major structural change.

In Li-ion, the lithium ions are incorporated into the crystal structure of the electrode material. This process involves a chemical reaction. The lithium-ions take time to diffuse into the bulk of electrodes. If the Li-ion battery is charged too quickly, this leads to defects in the crystalline structure and the deposition of lithium metal on the electrode in the form of lithium dendrite crystals. These lithium crystals can penetrate through the separator layer and cause a short circuit with the counter electrode, which could result in a thermal run-away and fire. Over-charging and deep discharging of Li-ion battery may lead to deterioration of the components.



# SUPERCAPACITORS

Supercapacitors, also known as ultra-capacitors, electrochemical capacitors or electrical double layer capacitors (EDLCs) are used for storing and delivering electrical energy several times faster than batteries. Figure 4 is a schematic of a supercapacitor.



However, EDLC's have had low energy densities so have not been used to store energy over a long period. By contrast, Li-ion batteries charge slowly but have higher energy densities. In supercapacitors, energy is stored at the surface of the electrodes electrostatically, by accumulation of charges. The ions from the electrolyte migrate to the electrodes, and accumulate at oppositely charged electrode surfaces during the charging process as shown in Figure 5.

The ions at the electrode/electrolyte interface form an electrical double layer during the charging process. The amount of energy that can be stored in a supercapacitor depends on the surface area of the electrode, in particular the electrode/electrolyte interfacial area, concentration and size of the ions and the operating voltage.

Most commercially-available supercapacitors are constructed by winding a pair of carbon coated aluminum foils with a separator sandwiched in between, and by filling the cell with an electrolyte. Activated carbon is often the active material and either acetonitrile or propylene carbonate containing tetraethyl ammonium tetrafluoroborate (TEA BF<sub>4</sub>) is the electrolyte. The operating voltage of the cell is up to 2.7V. The energy stored in a supercapacitor can be expressed as  $E = \frac{1}{2}CV^2$ , where E is the energy stored (in Joules), C is the capacitance of the cell (in Farads), and V is the operating voltage (in volts). The operating voltage is limited by the electrochemical window of the electrolyte. The electrochemical stability of TEABF<sub>4</sub> in acetonitrile (CH<sub>3</sub>CN) or in propylene carbonate is limited to below 3.0V [9-13].

Activated carbon electrodes have relatively large surface areas (1500m<sup>2</sup>/g to 2000m<sup>2</sup>/g) and inter-connected pore-structure. There are three types of pores:

- Macropores (>50nm);
- Mesopores (2-50nm); and
- Micropores (<2nm)

The micropores are important for electrical double layer formation and hence for energy storage. However, the presence of large diameter macropores and mesopores tends to reduce the electrode/electrolyte interfacial area and the specific capacitance of the electrodes.

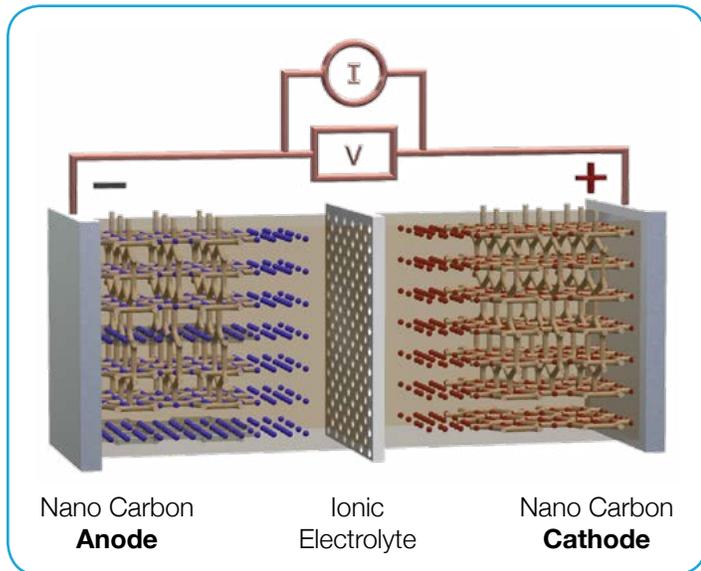
To produce an electrode coating, the activated carbon is blended with a conducting carbon and polymeric binders, which further reduces the active surface to some extent.

The energy densities of commercial supercapacitors are below 10Wh/kg as these cells are constructed using activated carbon and organic electrolytes.

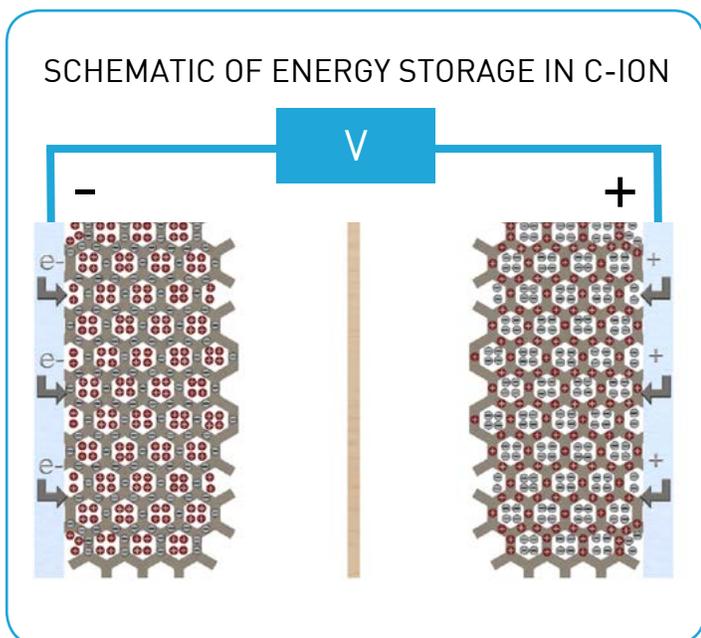


# CARBON-ION CELL

Recent advances in producing and fabricating a range of nano-structured carbons and ionic liquid based electrolytes have shown the feasibility of combining the power density of supercapacitors and the energy density of re-chargeable batteries. This cell is termed ‘Carbon-Ion cell’, or ‘C-Ion’, in this article to distinguish it from commercial supercapacitors. A schematic of a Carbon-Ion cell is shown in Figure 6.



These cells work in a very similar way to EDLC’s but use different carbon and electrolyte materials that are not only safer and easier to recycle at the end of life, but also enable the devices to operate at higher voltages resulting in higher energy densities. The energy storage in a C-Ion cell is schematically shown in Figure 7.



There is much interest in the potential for the nano-carbon material Graphene for energy storage because of its high surface area and high electrical conductivity. Graphene has a theoretical capacitance of 550F/g, and there has been progress in achieving part of this by addressing challenges such as stacking of layers [7]. The approaches include producing non-stacking 3D-graphene, making curved graphene platelets, using spacers such as carbon nano-tubes to preserve surface area and electrical conductivity, minimising defects and doping N, B and P to enhance the properties.

If the pore-size of nano-carbon is tuned to match the ion size, the capacitance can be considerably increased [8]. Nano-carbon with a pore size of 0.7nm showed the highest capacitance of 160F/g when 1-ethyl-3 methylimidazolium bis (trifluoro-methyl) sulfonyl imide (EMIM TFSI) was used as an electrolyte [8]. The size of the cation, EMIM+ and the anion, TFSI- were comparable to the pore diameter. The carbon electrode without matching the pore size to ion size showed a capacitance of only 100F/g. By increasing the operating voltage the energy density can be significantly improved. [8]

Some ionic liquids exhibit wide electrochemical stability and these are potential electrolytes to increase energy density of a C-Ion cell. They include EMIM TFSI, EMIM BF4, PYR14 TFSI, DEME BF4 etc.[8], Nano-carbons such as graphene, carbon nanotubes, carbon onions, carbide derived carbons and other forms of carbon have been used as active materials in supercapacitors and C-Ion cells. These experiments have shown promise for devices operating at 3.5V and beyond. Progress has been reported in improving the energy density by using nano-carbons and ionic liquids. [8,10]

Ionic liquids (IL) with a large electrochemical window tend to be several times more viscous compared to organic electrolytes. As a result, the ionic conductivity of an IL-based electrolyte is often low and this contributes to higher internal resistance in the cell as well as a compromise in power characteristics. During a heavy power demand the cell or a stack of cells will struggle to deliver huge power in a quick spurt of a few seconds.

The traditional supercapacitor design shown in Figure 3 envisages transport of charge carriers and ions in 2D geometry, and an ion in one of the electrodes located closely to the current collector has to migrate through the entire electrode and separator material to reach the other electrode to form the electrical double layer while being charged. The thickness of the carbon electrode is ~150 microns and the separator is ~20 microns. It is a tortuous path for the ion to move from one electrode to the other.



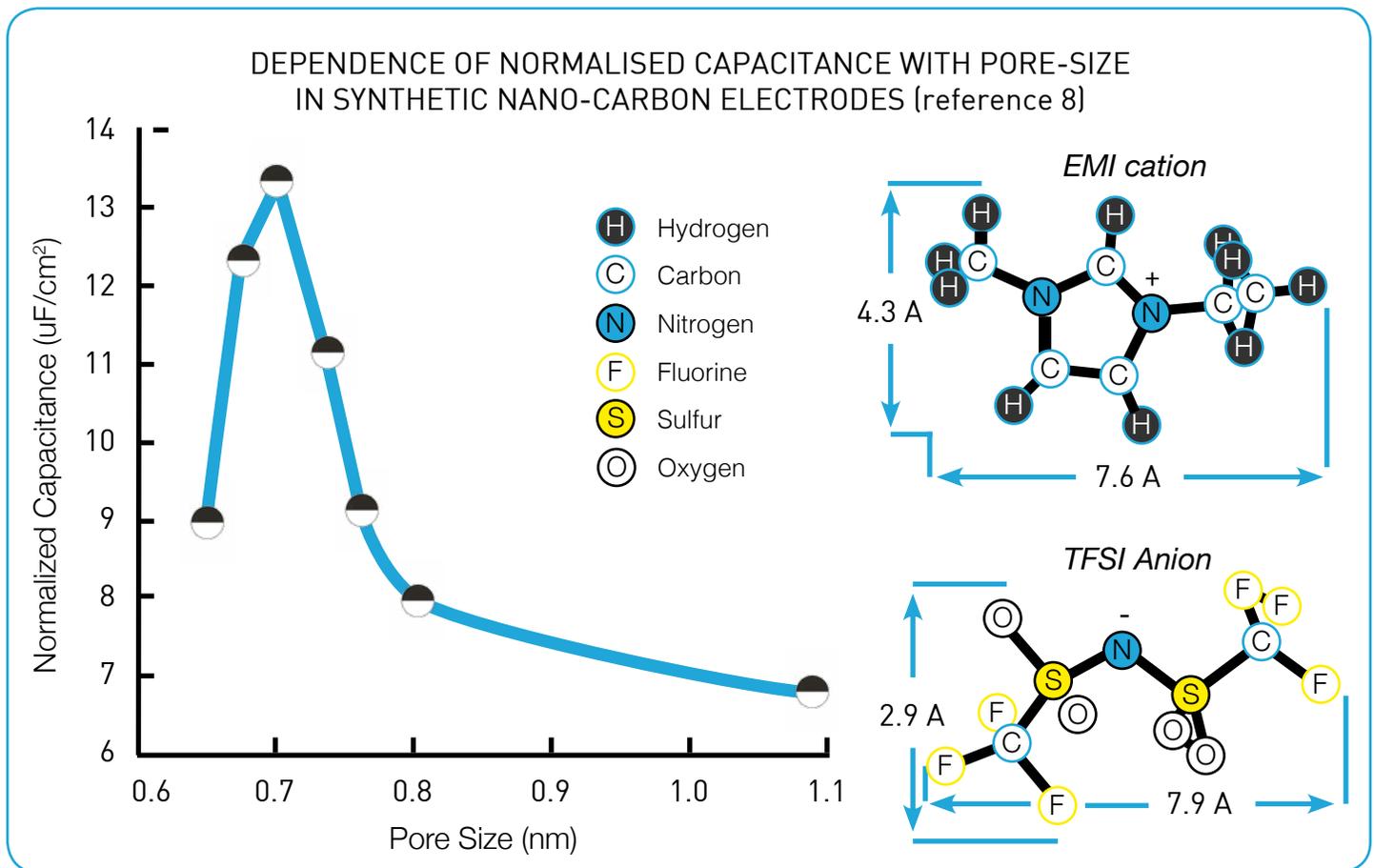
## INCREASING THE ENERGY DENSITY OF C-ION

By using synthetic carbons and nano-carbons it is possible to fabricate electrodes with controlled porosity. The amount of electrical energy that can be stored in a C-Ion cell is dependent on the surface area and electrical conductivity of the electrode, as well as the operating voltage of the electrolyte.

In recent years, advances in nano-structured carbons as electrodes and non-flammable ionic liquids as electrolytes have significantly enhanced the performance.

### NANO-CARBONS

Carbon materials have a high surface area and can be used as electrodes in electrochemical capacitors. Different types of nano-carbons in the form of powders, microspheres, fibres, foils and monoliths are commercially available. The physical and chemical properties of synthetic and nano-structured carbon materials such as graphene, single walled carbon nanotubes (SWCNT), multi-walled carbon nanotubes (MWCNT), fullerene and carbon onions are of interest as these materials have large surface area, unique nano-structures and the pore size in some of these materials is below one nanometer [7]. The specific capacitance of C-Ion cells can be increased by using nano-carbons with an optimum surface area and pore-structure.



Published results are summarized in Table 2. Nanoporous carbon has been reported with specific capacitance value as high as 284F/g (Farads per gram) [10] and 131Wh/kg. Activated carbon nanofibers [14] as electrode material exhibited a specific capacitance of about 60F/g. Exfoliated graphite oxide upon chemical reduction produced reduced graphene oxide and exhibited specific capacitance above 200F/g [11-12, 17-19]. Graphene based nanocomposites have also been explored as an active material. They have shown significantly increased capacitances. Functionalized graphene sheets were synthesized by thermal exfoliation method and achieved the specific capacitance value of 230F/g. [12]

## RELATIVE PERFORMANCE OF NANO-CARBON MATERIALS

ELECTRODE MATERIAL	PUBLISHED SPECIFIC CAPACITANCE (F/g) & ENERGY DENSITY (Wh/kg)	TEST CONFIGURATION	REF
Activated carbon (PICA CTIF)	125	Symmetrical cell	[9]
Activated carbon PICA A	115	Symmetrical cell	[9]
Carbide derived carbon	160	Symmetrical cell	[8]
Reduced graphene oxide (rGO)	284.5 (131 Wh/kg)	Symmetric cell	[10]
Thermally expanded graphite oxide	264	Symmetric cell	[17]
N-doped graphene	284	Symmetric cell	[18]
N-doped graphene & thermally expanded	270	Half-cell	[19]
Commercial RP-20 & steam activation	240	Half-cell	[11]
Thermal exfoliation of graphite oxide	230	Symmetrical cell	[12]
Curved graphene	154 (85.6 Wh/kg)	Symmetrical cell	[20]
Activated carbon fibre	60		[14]
Activated carbon & carbon nanotube	90		[15]
SiC-derived carbon	130		[16]

### IONIC LIQUIDS AS HIGH-VOLTAGE ELECTROLYTES

The energy density of a supercapacitor and a C-Ion cell can be increased by raising the operating voltage:  $E = \frac{1}{2}CV^2$ , where 'E' is the energy stored (Joules), 'C' is the capacitance (F) and 'V' is the operating voltage (V). Aqueous and organic electrolytes are stable up to ~1.2V and 3.0V respectively, and such lower operating voltage limits the amount of energy that can be stored in a supercapacitor.

Ionic liquids are a new class of electrolyte which are stable at higher operating voltages beyond 3.0V [7-8, 19-27]. Ionic liquids, having a wider electrochemical window, sufficient conductivity and lower viscosity, can be used as electrolytes in C-Ion cells. The physical and chemical properties of ionic liquids depend on the nature of cation, anion and the functional groups attached to these ions. Selected IL electrolytes show electrochemical stabilities up to 6V, and it is possible to tune the stability window by changing the cation-anion combinations. For the ionic liquids used as electrolytes, the conductivity values range from 1.0mS/cm to 10mS/cm [8]. Physical properties such as viscosity, conductivity and melting point can be varied by combining two or more ionic liquids.

### ELECTROCHEMICAL WINDOW OF ION ELECTROLYTES

IONIC LIQUIDS	ELECTROCHEMICAL WINDOW (V)	REF
EMIM BF4	4.6	[28]
EMIM TFSI	4.7	[28]
PrMIM TFSI	4.7	[28]
Bu3NMeTFSI	6.0	[28]
BuNMe3	5.3	[28]
BuMePyITFSI	5.7	[28]
PrMePiTFSI	5.7	[28]
DEME BF4	6.0	[29]
DEME TFSI	6.0	[29]

## RELATIVE PERFORMANCE OF IONIC LIQUIDS

IONIC LIQUID	CELL VOLTAGE (V)	ENERGY DENSITY (Wh/kg)	REF
EMIM BF4	4.0	85.6	[26]
EMIM BF4	4.0	136	[26]
EMIM BF4	4.0	131	[10]
EMIM TFSI	3.4	30	[27]
PYR14 TFSI	3.7	30	[27]
PYR12O1	4.0	40	[27]



## RECHARGEABLE BATTERIES OTHER THAN LI-ION

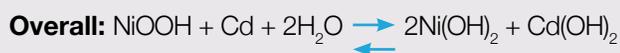
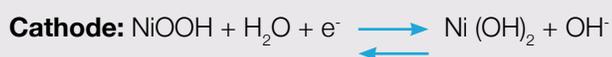
As well as Li-ion, there are two types of rechargeable battery in common use: **Nickel Cadmium (NiCd)** and **Nickel Metal Hydride (NiMH)**.

### NICKEL CADMIUM

There are in fact two types of re-chargeable nickel cadmium battery: one contains a 'sintered' electrode, the other type contains a 'non-sintered' electrode. The 'sintered' type suffers from 'memory effect' whereas the 'non-sintered' does not. The non-sintered electrode can increase the cell capacity by 40% (Sanyo KR-3UU: AA). The positive electrode is nickel and negative electrode is cadmium and the electrolyte is aqueous potassium hydroxide or sodium hydroxide. Energizer (Eveready Battery Company) introduced a 'Pasted negative electrode', with reduced memory effect, and Motorola developed a foam type nickel-cadmium battery to overcome this issue.

The nominal cell voltage is 1.2V, and the cycle life is from 500 to 1000 charge/discharge cycles. The energy density ranges from 45 to 80 Wh/kg and the cells can operate from -20°C to 65°C [31]. NiCd has the advantage of relatively high rate discharge, and the performance does not deteriorate at low- temperatures.

Nickel cadmium batteries are classified as 'hazardous waste': the cadmium is highly toxic and makes recycling them very difficult. [32-34]



### NICKEL METAL HYDRIDE

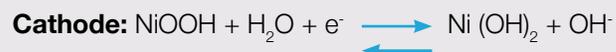
The nickel metal hydride battery was developed in the 1970s as an alternative to NiCd, removing the cadmium.

NiMH is similar to NiCd in both, the positive electrode is composed of nickel oxide (NiOOH) and a potassium or sodium hydroxide solution is used as the electrolyte.

The NiMH battery has a negative electrode which is a metal hydride alloy. This is an alloy which can store hydrogen.

Improved performance NiMH was introduced by Sanyo and Matsushita in the 1990's based on improved hydride alloys. Cell voltage is typically 1.2V.

Hydrogen gas dissociates to form absorbed hydrogen on the surface of the alloy. This then diffuses into the alloy and forms metal hydride. This is reversed during de-sorption.





## SUMMARY

Carbon-Ion cells are a new category of energy storage device that combine nano-carbons and ionic electrolytes to provide higher energy densities. They can also provide the rapid charging and long cycle life of supercapacitors, without any of the fire risk.

This technology can be used either on its own or in combination with Li-ion batteries for a range of rapid-charge applications, such as:

- Cordless power tools or domestic appliances
- In facilities such as emergency lights where its long life obviates the need to be replaced every few years
- ‘Peak Shaving’ in the solar energy infrastructure, mitigating the need for over-generation by generators
- In EV’s and hybrids for rapid energy capture through regenerative braking.

Ultimately it presents the opportunity for adoption in mobile phones, tablets, laptops and other portable devices.

### RELATIVE PERFORMANCE OF ENERGY STORAGE DEVICES

	LITHIUM (Li-ion)	SUPER-CAPACITORS (EDLC)	NICKEL CADMIUM (Ni-Cd)	NICKEL METAL HYDRIDE (NiMH)	CARBON-ION (C-Ion)
Energy Density	150-250 Wh/kg	5-10 Wh/kg	45-80 Wh/kg	60-120 Wh/kg	32-56 Wh/kg
Charging	Hours	Seconds	Hours	Hours	Seconds
Discharge rate	Slow	Fast	Medium	Slow	Fast
Cycle life	1,000	100,000	1,000	1,000	100,000
Safety	Flammable	Flammable	Flammable	Flammable	Non-flammable
Recycle	Poor	Poor	Poor	Poor	Good
Voltage	3.6V – 4.2V	2.7V – 3.0V	1.2V	1.2V	3V – 6V
Operating Temperature	-10°C to 60°C	-20°C to 60°C	-20°C to 65°C	-20°C to 65°C	-40°C to 80°C
Shipping	UN 38.3	UN 38.3	UN 38.3	UN 38.3	No restrictions



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## ABOUT ZAP&GO

Zap&Go was founded in 2013 in Oxford, UK with IP from the world leading University of Oxford in advanced nano carbon materials. These materials are used to replace the lithium commonly used in lithium-ion batteries with a safer, faster charging alternative. This means none of the fire risk associated with lithium batteries, and devices that charge much more quickly. Zap&Go has already developed a non-lithium powerbank mobile phone charger that charges in under 5 minutes and is planning further consumer and industrial product announcements that can reduce the charge time of everyday products from hours to seconds.

Zap&Go was the only energy storage company to make the Red Herring Global 100 list in 2016.

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