



Li-ion batteries and portable power source prospects for the next 5–10 years

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Abstract

This paper describes the possible evolution of Li-ion technology, and evaluates the expected improvements, arising from new materials to cell technology. New active materials under investigation and electrode process improvements may allow an ultimate final energy density of more than 500 Wh/l and 200 Wh/kg, in the next 5–10 years, while maintaining sufficient power densities. A new rechargeable battery technology cannot be foreseen today that surpasses this. The possible use of small fuel cells is also discussed. The only solution would be direct methanol fuel cell (DMFC) technology, providing that the remaining important technological issues are solved. The association with a rechargeable battery could provide an optimised energy + power, hybrid power source. Several simulated comparisons for small- to medium-sized power sources are described, between fuel cells and batteries. Hybrid Li-ion/DMFC is a good option for systems larger than 1 kWh. The hybrid concept of high-energy–low-power primary lithium (as the fuel) with high-power Li-ion (as the cell stack) is an already available, cost effective solution where long run times are required.

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1. Introduction

Making a prospective analysis of what could happen in the rechargeable battery field in the coming 5–10 years may appear quite easy when considering the slow evolution of this technology, compared to other fields, such as electronics. The Frenchman, Gaston Planté invented the lead acid battery at the end of 19th century, and more than 100 years later, this battery system is still the most produced all over the world, albeit with some improvements. By comparison, the electronic radio valve, which appeared much later, has been on museum shelves for a long time. The simple reason can be found in the chemical nature of batteries. They are centred on electrochemical reactions which involve materials, and the energy produced is directly proportional to the quantity of these reactant materials. “Miniaturisation” at constant energy is therefore impossible if we have stay with the same chemical reactants. Unfortunately, the possible suitable combinations are far from infinity, and improvements of only one order of magnitude are even unrealistic.

However, considering what has happened in the last 10 years may appear to invalidate this way of thinking. Indeed,

an apparently totally new battery concept appeared, known as Li-ion, which can be considered as a revolution in the battery field. In less than 5 years, this system has taken the largest part of small portable power sources, to the detriment of traditional Ni/Cd, or newly developed Ni/MH, which is an evolution of alkaline aqueous batteries.

Although this concept appeared rather suddenly, one must consider that this is in fact the result of many years of research activity. Rechargeable lithium has indeed been the goal of many research projects from the early beginning of “lithium battery history”, which started about 40 years ago [1]. The concept of the “rocking chair battery”, which is implemented here, was proposed a long time ago [2] to circumvent the poor reversibility of lithium metal. Alloys—which are presently re-emerging—were very soon proposed. Graphite, which is the actual negative electrode being used in lithium-ion batteries, was also investigated very early [3]. This latter option was unfortunately not seriously considered, probably for two simple reasons. Firstly, the results were disappointing, because solvent co-insertion with lithium induced graphite exfoliation, and material destruction. Secondly, the comparison of specific capacities, $\text{Li} = 3860 \text{ Ah/kg}$ and $\text{LiC}_6 = 370 \text{ Ah/kg}$ were not very motivating.

After years of intensive but rather unsuccessful research activity on the reversibility of lithium plating, a major door

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was opened with the discovery of the insertion of lithium ions into the amorphous carbon structure, without exfoliation. Associated with LiCoO_2 , an already known positive material studied with lithium metal, the battery system was born [4], which was practically developed and named “Li-ion” a few years later [5]. In spite of the lower specific capacity of the negative material compared to Li, reasonable energy density was obtained with the first prototypes, compared to rechargeable lithium, in which lithium metal was introduced in very large excess to get round the poor reversibility.

This short history is recounted to point to the possible evolution from now on. The chances of finding a practical new battery system with a significant higher energy density in the next 10 years are extremely small. There is simply no such concept being presently actively studied at research level, as was the case for many years for rechargeable lithium. Li-ion has however been in constant evolution since its beginning, and will continue to evolve. The purpose of this paper is to discuss the different ways of improvement, and extrapolate the possible evolution in the future.

The other system which is more and more cited as the next generation for portable equipment is the fuel cell. This however has a totally different nature to other power sources, with its own constraints. From a short review of the state of art, and anticipation of the future, it appears that an efficient system would be a hybrid, i.e. combining a fuel cell and a rechargeable battery as a primary power source. Fuel cells will have to compete with advanced primary batteries, in terms of performance-to-cost ratio.

2. Evolution of Li-ion technology

2.1. Materials

The nature of the active materials is of course of primary importance to the resulting cell energy density. Continuous research is being carried on new electroactive compounds, with improved properties. The main features of the active materials which determine cell energy are the number of electrons they can store per unit volume or weight (volumetric capacity or specific capacity), and the electrochemical potential they produce.

Using a cell construction model, it is possible to calculate the energy density as a function of the intrinsic material properties. Fig. 1 illustrates the impact of specific capacity of the materials on the complete cell specific energy, using an “average” design of Li-ion electrodes and cell stack technology, as can be found in most of the commercial cells today.

2.2. Positive materials

The most widespread positive material for Li-ion, lithium cobaltite LiCoO_2 , produces a very high potential (up to about

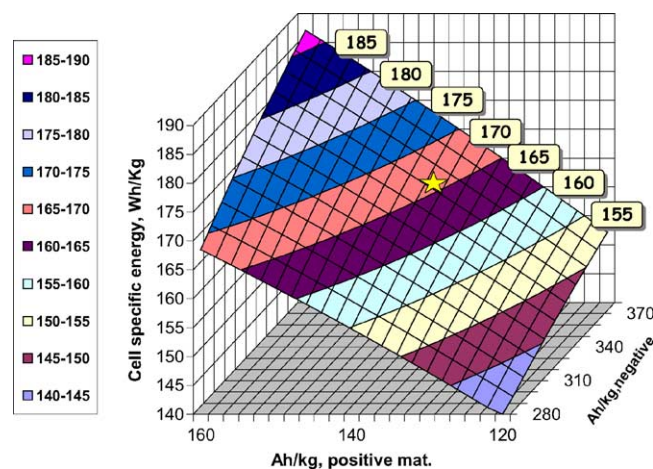


Fig. 1. Impact of material properties (negative and positive electrodes) on cell specific energy. Star indicates a LiCoO_2 /graphite example.

4.3 V versus Li/Li^+) when oxidised during charge. Discovered several years ago [6], it was nevertheless not industrially produced until this new battery system was launched in 1990. Thanks to the materials ability to release about half (0.5) a lithium atom per mole, and oxidise half of the cobalt to the tetravalent stage, the resulting specific capacity is close to 140 Ah/kg, and capacity density 690 Ah/l.

LiNiO_2 , a similar oxide using nickel, has a higher specific capacity (up to 200 Ah/kg during the first charge). However, this also already known material was much more difficult to synthesise, and had some drawbacks: less stable in the overcharge state, which increases the potential hazards on abuse, and its slightly lower voltage is detrimental to the application of mobile phones.

It is nevertheless cited as a future material for energy improvement, and has been largely improved in the last 5 years by introducing elements as substitutes to nickel in the crystal structure, such as cobalt, aluminium or manganese. Such compounds are now used in large configuration cells [6], where the cobalt price is prohibitive. They show improved stability and excellent cycle and shelf life.

The third candidate for a Li-ion positive is the spinel-manganese oxide LiMn_2O_4 . The main advantages are the expected lower cost and a better stability on overcharge, which explains why it is more common in large batteries. However, compared to the others it suffers from a smaller energy density and a lower chemical stability, inducing a shorter life, especially at high temperature.

It is expected that the current on going research will produce materials with improved energy. A particularly promising field is the so-called “5 V class materials”, increasing significantly the cell working voltage with no or small sacrifice in capacity. One of the most cited, is $\text{Li}[\text{Ni}_{1/2}\text{Mn}_{3/2}]\text{O}_4$, with an average working voltage of about 4.7 V versus a lithiated graphite negative electrode, while maintaining more than 120 Ah/kg. This is the best result published till now, representing an approximate 15% energy density increase at the cell level.

Some problems have still to be solved to make this new system practical. The most important is the electrolyte, which must be able to sustain this high oxidising potential which is not achieved by the electrolyte solutions known till now.

2.3. Negative materials

Moving from a lithium metal anode to a lithiated carbon LiC_6 as described in the introduction induced a penalty of about 15–20% in Wh/kg of complete cells. However, the much higher material density (2 versus 0.54) makes the Wh/l comparison much more favourable. If one takes into account that a large excess of Li metal is necessary to make a maximum of 200 cycles, the energy densities of both systems are essentially similar. In any case, the cyclability would be very much lower than the thousands of cycles that can be demonstrated in suitable Li-ion chemistry and design [6]. Moreover, the lower safety, linked to the much higher reactivity of Li metal, especially when cycled, remains a very difficult issue, unlikely to be solved. As a conclusion, there is little chance that Li metal will come back and give practical batteries with improved energy density.

Search for pure lithium substitution by different metallic alloys was soon explored, with little success, to solve the problem of dendrites and low reversibility. The fact that they sometimes have much higher specific and volumetric capacity than LiC_6 made them again attractive, and induced a renewed interest and research in this field. Table 1 describes the main properties of such compounds. The volumetric capacity can even be in some cases higher than pure lithium! However, the first problem to solve is the volume change between the charged and discharged states. These big volume changes produce enormous constraints on the metal grains, and induce heavy fragmentation, very detrimental for cyclability. By comparison, the volume change of lithiated graphite LiC_6 is only 10%. This is actually a key point in the Li-ion concept, because the solid electrolyte interface (SEI) produced on the negative interface by reaction with electrolyte, is not destroyed during cycling. If that was the case, the small lithium loss, which would be consumed at each cycle to repair it, would result in rapid capacity drop, and poor cycle life.

Recent developments concentrate on nanoparticles of amorphous alloys, which could solve the problem of fragmentation. In some cases, an inert matrix (like oxides) around the particles would sustain the volume variation. However, the much lower density make the compounds less attractive on a volumetric basis, and the problem of SEI layer stability on cycling remains, whatever the particle size.

Some numbers illustrate this: the lithium loss observed during the first charge of a Li-ion battery to create the SEI on the graphite interface is generally of the order of 10% of the capacity. If during cycling, 5% of this layer is destroyed, 0.5% of the capacity will be lost at each cycle for repairing, and 100 cycles will result in 50% capacity loss. In the same way, the use of nanomaterials increases drastically the interface area to be passivated by SEI, increasing proportionally the amount of lithium lost at the first cycle.

These facts show the difficulty to succeed in this area and enlighten the exceptional properties of graphite in this role. Discovery of a new electrolyte, thermodynamically stable at this low voltage, would drastically modify the data of the problem, but this is unlikely.

A second factor influencing the complete cell energy density is the working voltage of the negative electrode versus Li metal. In most of the cases, the working potential of these alternative anodes is significantly higher than Li, therefore the cell potential is consequently reduced, which reduces the energy density. This is also the case of non-crystalline or amorphous carbons which can accommodate larger quantities of lithium than pure graphite. Their higher voltages, sometimes higher than 1 V makes them practically unsuitable.

Again, a construction model helps to define the goal on single material properties to reach a given improvement over the existing system. Fig. 2 describes the evolution of the calculated specific energy of a Li-ion cell, as a function of negative material properties. The electrode and cell design correspond to the average Li-ion design. The material is supposed to have a high density of 5 g/cm^3 , and average working voltage versus lithium is 0.4 V. Calculations are made for several efficiencies at the first cycle (SEI formation). The horizontal line at 155 Wh/kg represents an average value for the present Li-ion design.

As can be seen from this figure, the goal to surpass graphite is quite high, illustrating the difficulty of improvements in this area.

Table 1
Comparison of some properties of lithium alloys

Alloy	Specific capacity (Ah/g)	Volumetric capacity (Ah/cm ³)	Volume at charged state (m ³ /Ah)	Volume discharged state (cm ³ /Ah)	Variation (%)
Li	3.861	2.06	0.485	–	–
$\text{Li}_{22}\text{Sn}_5$	0.790	2.023	0.494	0.138	259
$\text{Li}_{22}\text{Si}_5$	2.012	2.374	0.421	0.102	312
Li_3Sb	0.564	1.788	0.559	0.227	147
Li_3As	0.840	2.041	0.490	0.163	201
LiAl	0.790	1.383	0.723	0.373	94
LiC_6	0.339	0.760	1.316	1.195	10

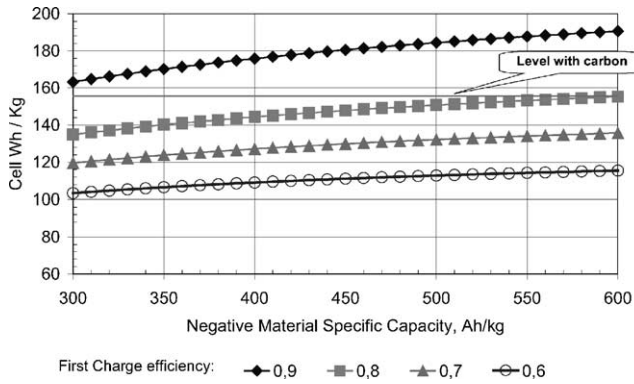


Fig. 2. Calculation of Li-ion cell energy density as a function of negative material properties.

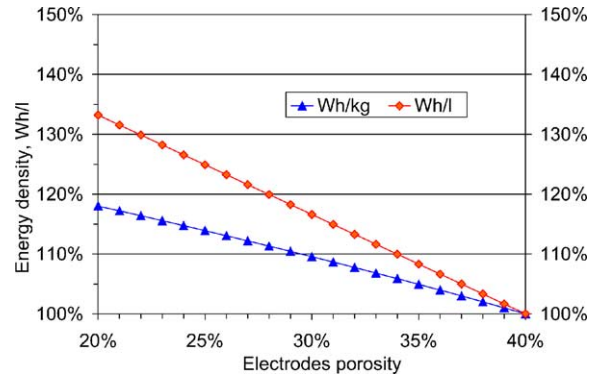


Fig. 4. Cell specific energy and energy density variation as a function of electrode porosities.

2.4. Cell technology, cell stack design

Beside the material properties, the electrodes and cell design are also important sources of improvement. In an average design, cell stack (electrodes + separator + electrolyte) occupy about 70–80% of the total volume (for medium-sized cells). The weight is about 80–85%.

These numbers have already been significantly improved since the introduction of Li-ion, but can still be further improved. It is anticipated however that, at best, an average of 10% in Wh/l could be gained. Wh/kg cannot be improved at this stage, except for the cells still using stainless steel cans, where aluminium can bring an improvement.

Fig. 3 represents the volume and weight distribution inside the cell stack. As it can be seen, the positive material weight is a major part of the total weight, while electrolyte occupies a very large part of the total volume.

As a general comment, the ratio between active components (positive and negative materials) and non-active is very dependant on the power required. A high-power design will necessitate larger electrode surfaces, more separator, electrolyte, and current collector.

Any improvement in the electrode designs, allowing better electrode kinetics, may result in a reduction of non-active components, for constant power. The clearest way for im-

provement is the electrolyte volume reduction, i.e. electrode porosity. The cell construction model can visualise the possible gains that can be obtained at the complete cell level, as illustrated in Fig. 4. In this case, the positive electrode and negative electrode are supposed to have the same porosity.

As could be anticipated, the influence is much more sensitive on volumetric energy. So, any improvement of power characteristics of the electrodes, through new electrolyte compositions, binder, active material physical properties, electrodes process, etc. can be translated into power, or energy increases. From the average present situation, an improvement of up to 20% can be expected in the coming years.

2.5. “Polymer” Li-ion cells

Essentially developed to produce thin small cells, this technology is in fact very similar to conventional Li-ion. The electrode designs are essentially the same, and cells contain as much liquid electrolyte as the usual design. The main difference is the use of a polymer to stick the electrodes together, producing an ionically conductive gel with electrolyte, and also playing the role of separator. By this mean, the self-sustaining stack can be placed in soft polymer packages, which facilitate the manufacture of thin

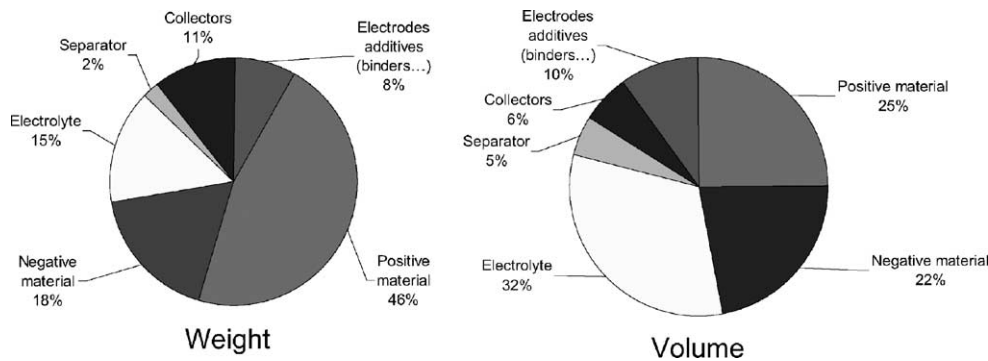


Fig. 3. Weight and volume distribution of cell stack components (“average Li-ion design”).

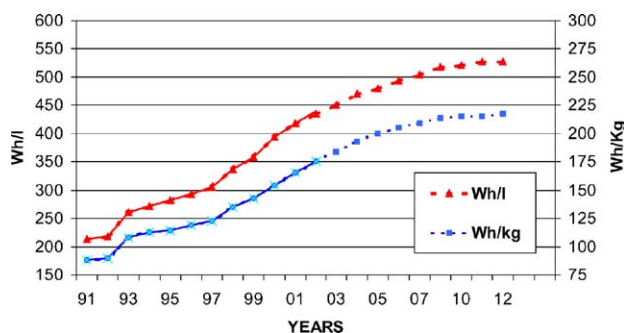


Fig. 5. Previous and expected evolution of energy density/specific energy of Li-ion batteries (low-power-high-energy design).

batteries. However, this design generally usually exhibits less volumetric energy in small cells, due to the size of the soft package sealing interface.

3. Anticipated evolution of Li-ion in the next decade

As a conclusion, one can expect a continuation of Li-ion improvements in the next decade, resulting from both material and electrode design improvements. In terms of specific energy or energy density of low-power cells, Fig. 5 illustrates what can be anticipated, by extrapolation of the past 10 years. More than 200 Wh/kg and 500 Wh/l should be attained in few years.

4. Portable fuel cells: the solution?

The fuel cell concept, mostly studied and developed for large energy systems for many years, is now more and more considered for small portable power sources. Some new concepts are even considering micro fuel cells, integrated in electronic chips. They are very often cited as the next future step for the high-energy portable power source. Several manufacturers announced recently the development of portable computers powered by small direct methanol fuel cells (DMFCs). Their operation range (about 4 h) is however not really more than can be achieved today with an appropriate Li-ion battery. Much longer operation is said to be expected in the future. Different from a rechargeable battery, the fuel cell does not store electricity but produces it irreversibly from chemicals, supplied from outside the electrochemical stack. The oxidising agent is atmospheric oxygen, and possible fuels are hydrogen gas, natural gas or liquid fuels such as methanol and petroleum.

There are several fuel cell concepts, the only one suitable for small systems, working at ambient temperature, is the proton-exchange membrane fuel cells (PEMFC).

The basic simple electrochemical reaction is the oxidation of hydrogen by oxygen, producing water, the proton ions H^+ being transported through the polymer proton-exchange membrane.

The overall simple reaction, $H_2 + (1/2)O_2 = H_2O$, produces a high theoretical energy, however about half of it is converted into heat. The electrochemical reaction necessitates a catalytic interface, mainly obtained with platinum.

The most developed design uses hydrogen gas as a fuel, either stored in a tank or produced in a reformer by thermal cracking of a liquid fuel. The second option is to use methanol directly in the direct methanol fuel cell.

The aim of this paper is not to describe in details the technologies, but to give some idea of the energy densities and specific energies which can be practically expected from the different options, in portable systems.

4.1. System architecture

A complete fuel cell system can be described with several basic components:

- (1) The electrochemical stack, which essentially includes the electrodes, the membrane, current collectors, channels to supply and evacuate fluids (hydrogen gas or methanol, air, water, etc.). This is the “heart” of the fuel cell. Its size is directly related to the required power.
- (2) The fuel tank, the volume is proportional to the required energy.
- (3) All the additional “non-active” items to supply and remove or treat fluids (reformer, pumps, fans, valves, humidity regulators, etc.) Also called “balance of plant”, this part is more or less proportional to the required power, but it has a minimum size and weight. This is obviously a critical point when the other parts are reduced, as is the case for portable fuel cells.

For this reason, the DMFC appears to be the only system which could provide a good solution.

4.2. The hydrogen options

Hydrogen can be supplied from:

- Liquid hydrogen: obviously not suitable for portable applications.
- High-pressure reservoirs (currently 300 bar, expected to reach 700 bar in the future). This solution is the simplest, but appears to be not efficient for a miniature cell in terms of energy density. For example, an existing 50 W/12 V system [7], has a stack weight of 2.9 kg, with a volume of 4.3 l. A 900 Wh high-pressure hydrogen bottle, that when empty weighs 5.2 kg results in a total system weighing about 8.81 kg, giving 173 Wh/kg and 102 Wh/l system. Despite the fact that this medium-sized system is not optimised, one can anticipate that it will be extremely difficult for a smaller sized fuel cell to reach the desired level of energy density, to compete with Li-ion, either in terms of Wh/l or Wh/kg. Comparison with lithium primary batteries will be even more difficult.

- Chemicals like NaBH_4 are options being studied to improve the energy density of the fuel tank (3.9 kg expected instead of 5.2 kg in the previous example). This however needs a reformer to produce hydrogen from the thermal degradation of the solution in water, and recycling of the water produced in the cell stack, to reach better efficiency. If this concept could be applied to a sufficiently large volume cell, its application to small portable cells will be less efficient.
- Metal hydrides: very limiting for specific energy, because of the metal weight.
- Liquid fuels such as methanol through a reformer, at high temperature on a platinum catalyst: this option is currently preferred for the large fuel cells for EV, but would be very difficult to apply to portable fuel cells, needing sophisticated “micro reformers”.

4.3. Direct methanol option

In this case, methanol is oxidised at the negative electrode surface, to produce carbon dioxide and protons, which move through the membrane to the positive electrode, where they are converted into water by oxidation with atmospheric oxygen.

The general equation is: $\text{CH}_3\text{OH} + (3/2)\text{O}_2 \rightarrow 2\text{H}_2\text{O} + \text{CO}_2$.

Water is involved at the negative electrode: $\text{CH}_3\text{OH} + \text{H}_2\text{O} \rightarrow 6\text{H}^+ + 6\text{e}^- + \text{CO}_2$, and is produced at the positive through: $6\text{H}^+ + 6\text{e}^- + (3/2)\text{O}_2 \rightarrow 3\text{H}_2\text{O}$.

Therefore, an efficient system should recycle part of the water produced at the positive electrode to supply the negative compartment.

This is, in fact, on theoretical basis, a promising feature for building small portable fuel cells. The theoretical energy density of the fuel is very high (6 kWh/kg and 5 kWh/l, based on pure methanol). The direct introduction of the liquid into the cell in contact with the catalytic surface of the positive electrode may appear very simple (like a conventional battery) with less burden attached to the electrochemical stack than for the technology previously described.

There are however several serious drawbacks, which make the perspective much less encouraging:

- The actual practical electrochemical energy recoverable is much lower than the theoretical value. From the total energy, less than about 30% can be expected as electricity, the rest being converted into heat. The heat produced has to be dissipated at the system level, which can be a strong disadvantage for some compact portable systems.
- This still high energy density (about 1.8 kWh/kg and 1.7 kWh/l), must be “diluted”, as pure methanol cannot be used. Diluted solutions in water are actually used, in concentrations of approximately 10%. The main reason is the “cross-over” phenomenon, which is still the most important fundamental problem to solve. By this mechanism, methanol molecules diffuse through the membrane,

and are directly oxidised by oxygen on the positive electrode catalytic surface. This has two negative effects: self discharge of methanol, not producing electricity but additional heat, and—even more detrimental—drastic reduction of the positive electrode voltage. This is like a “chemical short circuit”.

As a result, the present practical fuel energy density is much reduced (less than 170 Wh/l, if 10% solution is used). Practically, methanol must be diluted with recycled water produced by the reaction, before introduction in the cell. This can be achieved in a medium-sized system, but would be much more difficult to apply to a small system.

- The power density available from the methanol oxidation at the negative electrode is much lower than for hydrogen oxidation (about 1/10). This has two consequences:
 - More platinum + ruthenium catalysts are needed (10× compared to hydrogen), with the associated cost problem.
 - The cell stack dimension must be proportionally increased. DMFC is a low-power-density fuel cell.
- Poisoning of the catalyst by CO.
- Beside these items specific to DMFC, the membrane cost and life time is still an issue.

It is very difficult today to figure out from the literature what energy and power density can be achieved in the future with portable DMFC systems.

From the above consideration, it appears that fuel cells will hardly compete with small portable batteries. Medium-sized systems can be effective for transportable power and some products are at an early commercial stage [9]. For example, a 25 W/1.7 kWh size unit exhibits up to 170 Wh/kg, but less than 100 Wh/l because the system is not designed to optimise volume. Such systems can be used, for example, in remote areas as battery chargers. From anticipated specifications of products under development, an optimised 25 W cell designed to reduce volume would produce 340 Wh/kg and 245 Wh/l for a large 1.7 kWh unit. The goal of a CECOM development program (DARPA Palm Power) is a 20 W/1 kWh DMFC unit producing 500 Wh/l and 750 Wh/kg [8]. This can be considered as an “ideal” DMFC, and could be achieved by technical breakthroughs as described above.

However, when considering a smaller unit (less energy required, with the same power), energy density drastically decreases. The performance of the optimised 25 W cell mentioned above falls down to 40 Wh/kg and 35 Wh/l for a “small” 3.5 dm³/120 Wh system, i.e. allowing a continuous running time of about 6 h at 20 W average.

As an illustration, Figs. 6–8 show comparative simulations of several solutions for a 25 W average/100 W peak power source. Total weight and volume are calculated, as a function of the required energy, or operating time.

As mentioned earlier, fuel cells should be considered as a two-component system: power is generated by the electrochemical stack, energy by the fuel reservoir. The required

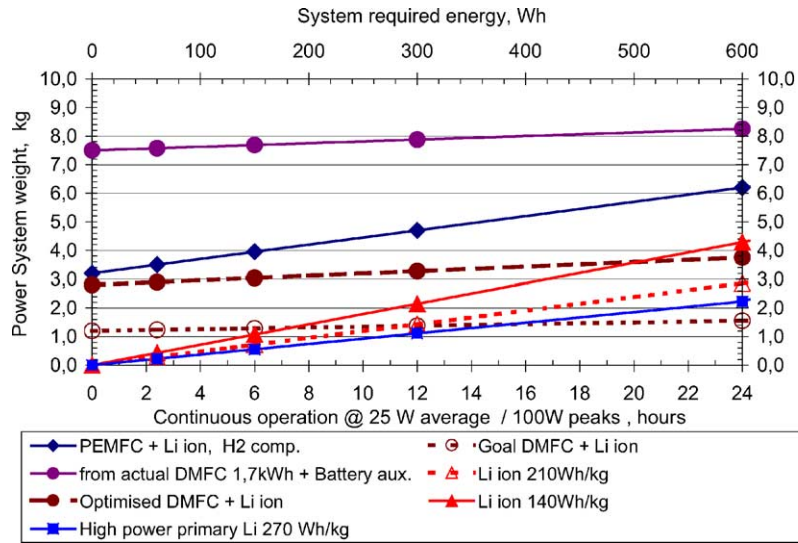


Fig. 6. Weight comparison of several solutions for a 25 W portable power sources as a function of required energy, for up to 1 day of operation.

power will determine the size of the stack (and its cost.). Therefore, when calculating the size of the system, as a function of energy needed E , the total volume (or weight) follows a function $V = V_0 + kE$, where V_0 is the volume of the cell stack, depending on power requested including appropriate additional managing equipment, and k the inverse of fuel tank energy density (l/Wh).

Because of the low power density of a DMFC cell stack, a hybrid system with a high-power-density battery to provide the power peaks significantly increases the overall energy density or specific energy of the system, and reduces its cost. So rather than being competitors, fuel cells and rechargeable batteries are actually complementary. This reasoning is also true for pure hydrogen fuel cell stacks, and large systems. For a complete system, the fuel cell stack will be sized as a

function of the required average continuous power, and the additional high-power battery will be sized to provide peak power. Then the size of the fuel tank(s) will determine the total available energy.

In the simulations, a small high-power Li-ion battery (200 g/80 cm³) provides the 100 W peaks. Fig. 6 describes the weight of small systems, up to 1 day of operation. The values calculated from the actual existing 25 W/1.7 kWh DMFC are much heavier than the other options. The figure also shows clearly that the PEMFC with compressed hydrogen cannot be competitive. The weight of the present rechargeable Li-ion is always less than an expected optimised DMFC. Even the goal for the “ideal” DMFC is not competitive until 12 h of continuous operation versus an optimised 200 Wh/kg Li-ion. The figure also displays the

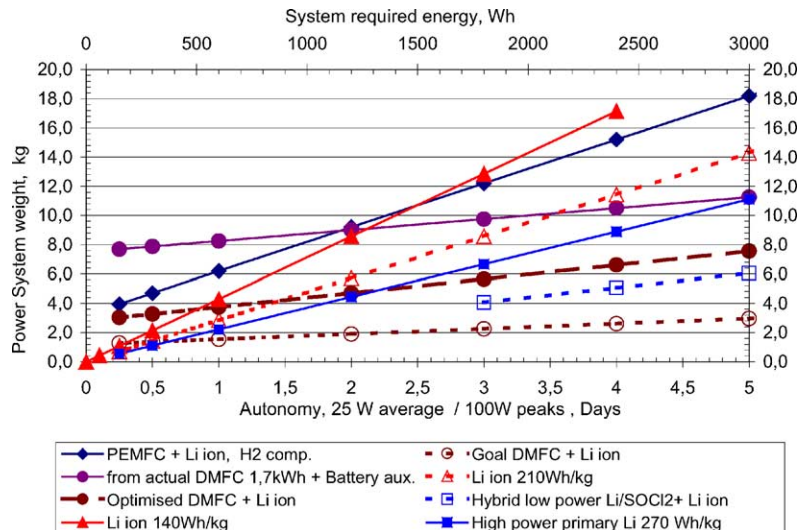


Fig. 7. Weight comparison of several solutions for a 25 W portable power source as a function of required energy, up to 3 kWh, or 5 days of operation.

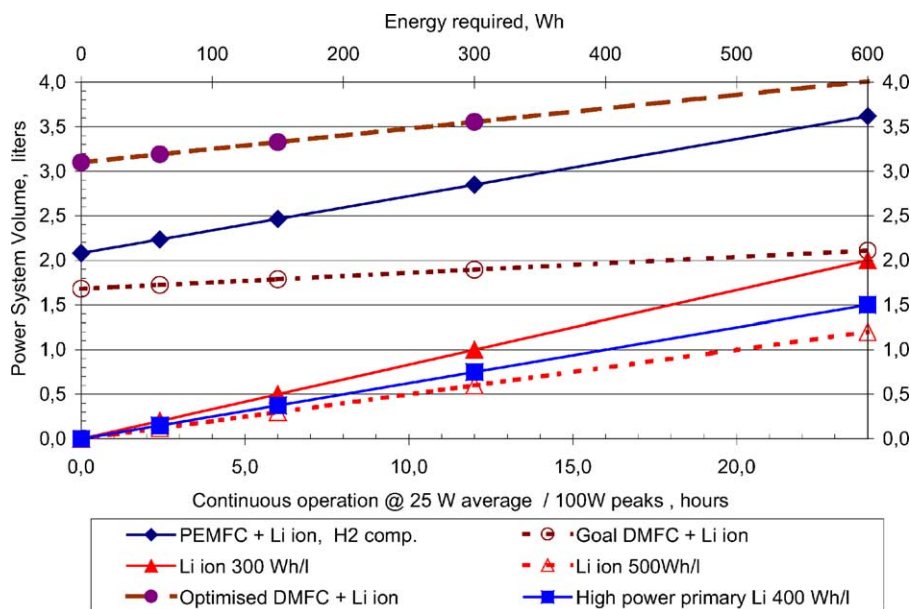


Fig. 8. Volume comparison of several solutions for a 25 W portable power source as a function of required energy, up to 1 day of operation.

performance obtained with an existing high-power lithium primary battery, where the weight is definitely the lowest of the options for such applications.

Possible advantages of a fuel cell will consequently be found in power source systems requiring long operation time. Fig. 7 describes such systems with the same power as in the previous example, but providing up to 5 days of continuous operation.

It can be seen that the goal for the “ideal” DMFC is actually the best technical solution. The expected medium term optimised DMFC, as described above, is better than an optimised future Li-ion battery at 200 Wh/kg if it is larger than about 1 kWh. The comparison with primary lithium extends this limit to 1.5 kWh.

Interestingly, the hybrid concept has been applied to a high-specific-energy–low-rate primary lithium associated with a high-power lithium ion to provide the peak power. The simulation is made using low-rate bobbin-type Li/SOCl₂ cell, which are the highest energy density cell available today (up to 700 Wh/l and 530 Wh/kg), higher than the frequently cited zinc air system (practical, for a 500 Ah cells: 380 Wh/l and 300 Wh/kg). It can be seen that this option is very efficient, better than the mid term optimised DMFC. This is however valid for large systems only, because the primary source must be able to provide at least the average continuous power required, for example, about 2 kWh at 25 W, which represents a C/80 discharge. This is a very attractive option, which already exists.

When the size of the power source is more important than the weight, fuel cell is less favourable. This is illustrated in Fig. 8, for less than a 600 Wh requirement, where the battery options are always better, even compared to the “ideal” DMFC. Larger systems become more favourable for

fuel cells, on a volume basis, where there is more competition with conventional power sources. In particular, the hybrid primary/rechargeable lithium appears to be close to an ideally optimised DMFC, up to 2.2 kWh, or 4 days of operation.

5. Conclusions

No revolution is expected for portable battery systems in the next 5–10 years. Energy density and specific energy of Li-ion, presently the best technical solution, will still increase but will rapidly reach a limit. Portable fuel cells using the DMFC concept can be theoretically the best future solution for medium-sized systems, larger than at least 0.5 kWh. There are however a number of serious problems to be overcome, and the commercialisation of a competitive viable system versus an improved battery cannot be expected before at least 5 years. To overcome the low power density of DMFC system, hybrid systems using an auxiliary high-power-density Li-ion battery will be the best option in most cases. When long term operation is required for medium power systems, a hybrid system primary lithium/Li-ion appears to be a very attractive option that is available today.

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