



# Lithium polymer batteries and proton exchange membrane fuel cells as energy sources in hydrogen electric vehicles

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## ARTICLE INFO

### Article history:

Received 1 October 2009  
 Received in revised form  
 15 December 2009  
 Accepted 16 December 2009  
 Available online 22 December 2009

### Keywords:

Lithium batteries  
 PEM fuel cells  
 Hydrogen electric vehicles  
 Energy management

## ABSTRACT

This paper deals with the application of lithium ion polymer batteries as electric energy storage systems for hydrogen fuel cell power trains. The experimental study was firstly effected in steady state conditions, to evidence the basic features of these systems in view of their application in the automotive field, in particular charge–discharge experiments were carried at different rates (varying the current between 8 and 100 A). A comparison with conventional lead acid batteries evidenced the superior features of lithium systems in terms of both higher discharge rate capability and minor resistance in charge mode. Dynamic experiments were carried out on the overall power train equipped with PEM fuel cell stack (2 kW) and lithium batteries (47.5 V, 40 Ah) on the European R47 driving cycle. The usage of lithium ion polymer batteries permitted to follow the high dynamic requirement of this cycle in hard hybrid configuration, with a hydrogen consumption reduction of about 6% with respect to the same power train equipped with lead acid batteries.

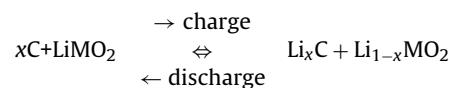
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## 1. Introduction

Hydrogen fuel cell propulsion systems are surely advantageous with respect to internal combustion engines, and at the present state of development of batteries they are also competitive with electric vehicles. While the internal combustion engines are not convenient in terms of efficiency and emissions (NO<sub>x</sub> would be present also if hydrogen was used as fuel) the electric vehicles suffer of the well known problems associated to the presence of batteries (weight, size and limited driving range). The utilization of hydrogen fuel cells on electric vehicles would permit to reconsider the role of the electric energy storage systems, to guarantee the crucial characteristic of zero emission and to increase the driving range thanks to the presence of the fuel on board, like for the conventional vehicles [1,2].

In recent years Li based batteries have been object of an increasing interest as electric energy storage systems characterized by high energy/power density, which is the most crucial requirement of electric vehicles. Lithium metal is attractive as battery anode material mainly due to its lightness and high voltage. Of course, the use of a lithium based anode in these systems implies some concerns of safety hazard, due to the high reactivity of the metal. For this reason, in the so-called lithium-ion batteries, both positive and negative electrodes employ lithium “host” compounds, where an interca-

lation process occurs, i.e. lithium ions are reversibly removed or inserted without a significant structural change to the host. The negative material is based on graphitic carbon, while the positive one is realized by using a metal oxide as lithium source compound, of the LiMO<sub>2</sub> or LiMO<sub>4</sub> type (M = Co, Ni, or Mn). In these systems the lithium ion conducting electrolyte is based on a solution of a lithium salt in organic solvents [3]. The resulting electrochemical reaction is the following:



which involves a cyclic transfer of lithium ions from the cathode (the lithium source) to the graphite anode, with no metallic lithium present in the system. This type of Li battery has already widely diffused in the electronic consumer market, however for automotive applications the presence of a liquid electrolyte is not considered the best solution in terms of safety, then for this type of utilization the so-called lithium polymer batteries appear more convenient. They are based on a polymeric electrolyte which permits the transfer of lithium ions between the electrodes [4]. The anode can be composed either of a lithium metal foil (in this case the device is known as lithium metal polymer battery) or of lithium supported on carbon (lithium ion polymer battery), while the cathode is constituted by an oxide of lithium and other metals, of the same type used in lithium-ion batteries, in which the lithium reversible intercalation can occur. For lithium metal polymer batteries the overall cycling process involves the lithium stripping-deposition

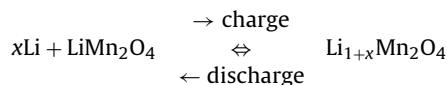
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**Table 1**  
Open circuit voltage (OCV) and specific energy values for different electrochemical pairs<sup>a</sup>.

Pair/fuel	Theoretical OCV (V)	Actual OCV (V)	Theoretical specific energy (Wh kg <sup>-1</sup> )	Actual specific energy (Wh kg <sup>-1</sup> )
Pb/acid	2.1	2.0	252	30–45
Ni/Cd	1.35	1.2	244	40–51
NiMH	1.35	1.2	206	50
Li/ion	4.1	4.1	410	150
Li/polymer (MnO <sub>2</sub> )	3.5	3.0	1000	120
USABC	–	–	–	200
Gasoline	–	–	–	13,000

<sup>a</sup> Adapted from Ref. [5].

at the anode, and the deintercalation–intercalation at the anode, according to the following electrochemical reaction, written for a Mn based cathode:



The absence of liquid phases facilitates the construction of leak-proof and light-weight containers, which represents an additional advantage for automotive applications.

Recent developments in this field have been focused on the possibility to reach very high energy and power densities by using new types of anode and cathode. Metals and semiconductors, such as Al, Si, Sn, Bi, have been considered for their capacity to form alloys with lithium, which are characterized by a theoretical charge capacity very higher than traditional carbon materials (in particular, a Si–Li alloy presents a theoretical specific capacity of 4200 mAh g<sup>-1</sup>, to be compared with 371 mAh g<sup>-1</sup> of graphite [5,6]). However, the large volume change associated with the formation of the alloys (by lithium intercalation/deintercalation) rapidly leads to electrode pulverization, strongly limiting the cycling capability of the battery [6]. To overcome this type of limitation different solutions are under study, from reduction of metal particle size down to nanoscale [7,8] to utilization of composite materials (in which an inactive component added to the active metal acts as a buffer for volume variations) [9,10] or metal hydrides as anode [11]. The researches about the cathode of lithium ion batteries are intensively oriented on high voltage spinels and high capacity layered lithium metal oxides [12–15].

Even with the current technology the lithium polymer batteries represent the state of the art in the field of electric energy storage systems, since they are characterized by very interesting values of the basic electrochemical parameters, as reported in Table 1 in comparison with other batteries today available. The theoretical open circuit voltages (OCV) represent the electrochemical potential of the single cell reaction, while the theoretical specific energies are calculated applying the Faraday law. When a practical battery is assembled, the weight of inactive components necessary to its construction increases the total weight of the battery without contributing to energy, then the actual values for both parameters are also reported in Table 1 to take into account the characteristics of real systems and the possible variations among different manufacturers [16].

In different countries various organizations have been funded with the aim of promoting research activities in the field of electric energy storage systems for automotive applications, and of individuating goals to be met by the innovative batteries in order to fulfil the requirements of electric vehicles acceptable by the market. In United States the Advanced Battery Consortium (USABC), founded in 1995 by Ford, General Motors and Chrysler in cooperation with Department of Energy (DOE), have proposed that a battery for electric vehicles should be able to store at least 200 Wh kg<sup>-1</sup> to afford the vehicle an acceptable driving range, and this value is generally accepted as benchmark for battery development programs

worldwide. For comparison the specific energy value established by USABC for future acceptable batteries and the value for gasoline are also reported in Table 1.

The actual specific energy data evidence the fundamental problem of today battery vehicles, i.e. the driving range connected to limited specific energy of the currently available batteries (maximum 50 Wh kg<sup>-1</sup> for NiMH, and lower values for Pb/acid). On the other hand, also with lithium systems, whose higher voltage would reduce the number of cell in a battery pack by a factor of at least 2, storage capabilities compatible with transportation applications are not reached.

A fuel cell propulsion system can be developed using the fuel cells as unique power source, without the support of batteries, or in hybrid configuration, where the storage systems play an important role in power supplying. Even if the driving range of a fuel cell vehicle depends on the quantity of hydrogen stored on board, the possibility to adopt high performance batteries, such as lithium based systems, is of great interest for all hybrid solutions fuel cells/batteries which are considered necessary for application of hydrogen to electric traction [17].

For this paper a lithium ion polymer battery pack (12 V, 40 Ah) was characterized in charge–discharge test cycles at different constant current values, in order to evaluate its performance in comparison with Pb acid batteries undergone to the same test procedures. Then two lithium ion polymer batteries (48 V, 20 Ah) were integrated in a fuel cell power train for moped application installed on a dynamic tests bench, and some preliminary tests were carried out on the European R47 driving cycle in order to have indications about fuel economy and dynamic behaviour issues associated to the utilization of this type of batteries in hybrid configuration with hydrogen fuel cells.

## 2. Experimental

The lithium based storage systems used in the present work are of the type known as lithium ion polymer batteries, and are constituted of a graphite based anode, a Li(NiCoMn)O<sub>2</sub> based cathode and a Li<sup>+</sup> conducting gel polymer electrolyte as separator. Their main characteristics and recommended operative conditions are reported in Table 2 for a single module. The characterization tests were effected on a battery pack constituted by a series of 4 elements, each of them composed by 2 single modules connected in parallel (for a total of 8 modules), utilizing a battery tester whose technical characteristics are reported in Table 3. This equipment adopted an acquisition system able to monitor the electric parameters (current and voltage) and battery temperature during the run, and to program different types of test procedure.

The lithium battery pack was tested in charge/discharge cycles respecting the recommended values of maximum voltage in charging (16.6 V) and minimum voltage in discharging (10 V), in order to avoid alteration of battery cycle life guaranteed by the supplier, while the values of charging/discharging current were varied in the range 8–100 A (from C/5 to 2.5C, where C is the battery capacity). Two types of test cycles were adopted: (1) the battery pack was fully

**Table 2**  
Main characteristics and recommended operative conditions of lithium ion polymer modules<sup>a</sup>.

Characteristic	Value
Length	216 mm
Width	130 mm
Thickness	7.2 mm
Weight	410 g
Nominal voltage	3.65 V
Nominal capacity	20 Ah
AC impedance (1 kHz)	<3 mΩ
Specific energy	180 Wh kg <sup>-1</sup>
Energy density	365 Wh l <sup>-1</sup>
Maximum charge voltage	4.15
Lower discharge voltage	2.5
Temperature range	-30/+50 °C

<sup>a</sup> Data provided by the manufacturer.

charged or discharged setting a constant current value, in particular the charge was carried out until the power entering the battery decreased under 10 W, while the discharge was always taken down to the lowest voltage level recommended by the manufacturer, i.e. 100% of depth of discharge (DOD, defined as the level to which battery voltage is taken during discharge), when the run was stopped; (2) the battery was firstly fully charged at C/2 until the power entering the battery decreased under 10 W, then was discharged down to about 50% of its maximum state of charge (SOC), after that it was undergone at a sequence of 200 discharging/charging steps characterized by different values of constant current (2.5C in discharging and C/2 in charging), finally the battery was completely discharged (100% DOD). At the end of both types of cycles the battery capacity and the energy transferred in charge or discharge mode were determined, while the ratio of Wh discharged (100% DOD) to Wh supplied by the tester was assumed as a measure of the dynamic behaviour of the battery in a specific test procedure. Similar tests were also conducted on Pb acid batteries of the same type of those already used in previous experiments on the overall fuel cell power train [18].

The experimental tests with the fuel cell power train were effected using two lithium battery packs connected in parallel, each constituted by 13 modules connected in series (47.5 V, 20 Ah). The total capacity of lithium batteries resulted 40 Ah, comparable to that used in tests carried out with Pb acid batteries as reported in [18]. The detailed characteristics of the overall fuel cell power train are also described in the reference [18], and summarized in Table 4. It was constituted by fuel cell system, DC–DC converter, electrical energy storage system, electrical drive, data acquisition systems. The fuel cell system (FCS), was based on a 2.5 kW PEM stack fuelled with compressed pure hydrogen. A LAFERT brushless engine of 3.7 kW maximum power, of the same typology of that installed on electrical commercial scooters and equipped with a

**Table 3**  
Main characteristics of the battery tester used for characterization of lithium and lead battery packs.

Characteristic	Value
AC input	3 × 400 V 50 Hz
DC voltage	Up to 18 V
DC current	Up to 100 A in charge or discharge mode
Duty cycle	100% in charge or discharge at maximum current rate
Output ripple	Negligible
Efficiency	0.77
Power factor	0.9
Transition time from charging to discharging	130 ms

**Table 4**  
Main characteristics of the fuel cell power train<sup>a</sup>.

Fuel cell system	
FCS electric output	Max 2 kW after DC–DC converter
FCS dynamic	Max change rate 500 W s <sup>-1</sup>
Electric drive	
Type	Brushless
Maximum power	3.7 kW
Rated current	32 A
Maximum current	100 A
Link voltage	48 V
Motor voltage	35 V
Rated speed	3000 rpm
Maximum speed	6000 rpm
DC–DC converter	
Maximum inlet voltage	34 V
Minimum inlet voltage	19 V
Rated inlet voltage	24 V
Rated output voltage	48 V
Rated power	2.8 kW
Rated efficiency	86%

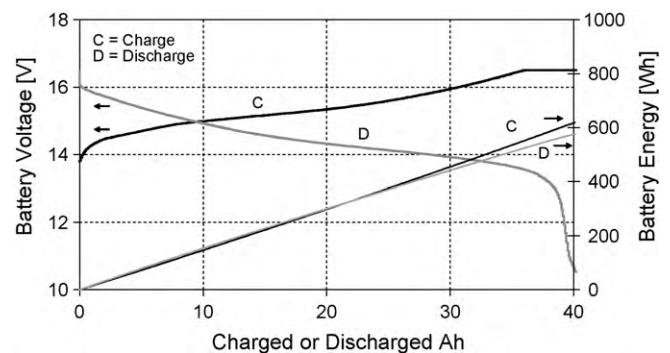
<sup>a</sup> Adapted from Ref. [7].

controlled inverter, was used. The DC–DC converter was necessary to match the stack output voltage to that required by the engine, and permitted to implement the control strategies corresponding to different hybrid configurations. The propulsion system was coupled to a braking electrical machine able to effect different driving cycles by a control software specifically developed. A preliminary test on the overall power train equipped with lithium ion polymer battery packs as electric energy storage system was effected on the European R47 driving cycle. A hard hybrid configuration was adopted as control strategy, setting the power supplied by the fuel cell system at a constant value corresponding to the average power required by the driving cycle, as measured at the DC–DC converter outlet.

During all tests the lithium battery packs were air cooled to hold the battery temperature under the recommended value of 50 °C.

### 3. Results and discussion

The results of the characterization effected on the lithium ion polymer battery pack (12 V, 40 Ah nominal capacity) are reported in Figs. 1–3. In particular, the voltage and energy curves during charge–discharge (DOD 100%) cycles for three different constant current values (20, 80 and 100 A) are shown as function of battery capacity. The Ah values reported in the abscissa were measured at battery terminals, and refer to the current fed to the battery during the charge phase, or that released by the battery during the discharge. During charge phases the typical voltage profiles



**Fig. 1.** Characterization test on lithium ion polymer battery pack (12 V, 40 Ah). Voltage and energy versus charged or discharged Ah at 20 A (C/2) during test cycle 1.

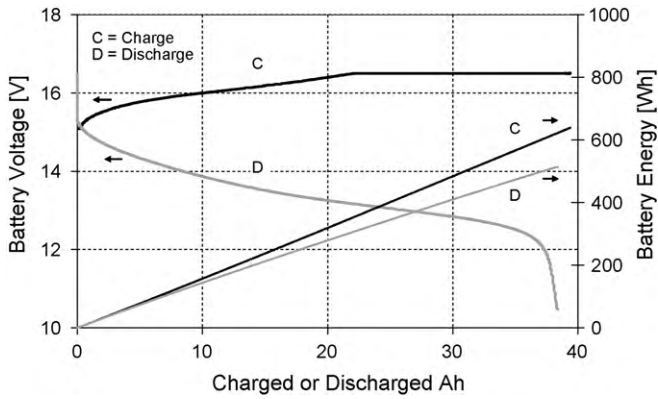


Fig. 2. Characterization test on lithium ion polymer battery pack (12 V, 40 Ah). Voltage and energy versus charged or discharged Ah at 80 A (2C) during test cycle 1.

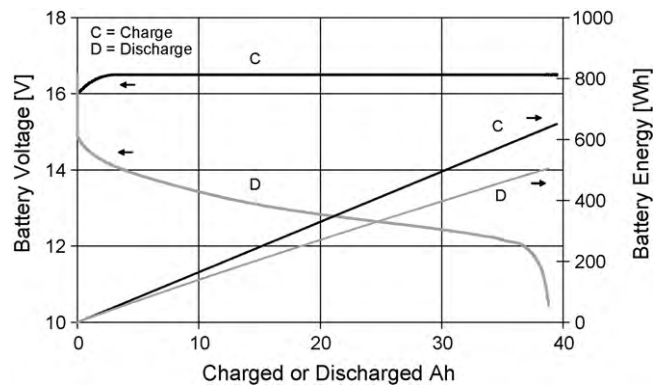


Fig. 3. Characterization test on lithium ion polymer battery pack (12 V, 40 Ah). Voltage and energy versus charged or discharged Ah at 80 A (2.5C) during test cycle 1.

versus capacity were observed, in particular the voltage values rapidly increased at the beginning of the charge phase, then more slowly reached the maximum value permitted (16.6V). The voltage increase rate depended on the charging current imposed, and was obviously higher at 100 A, in particular the maximum voltage was reached at 36, 22 and 2.5 Ah for 20, 80 and 100 A as charging current, respectively. The energy curves show that 630 Wh was fed to the battery at the lowest charging current before maximum voltage was reached, while 640 and 650 Wh was supplied to the battery at higher charging current values of 80 and 100 A. During the discharge phase down to DOD 100% (10V) the voltage profile decreased very fast at the beginning of the test, and this behaviour was particularly evident at 100 A when the voltage decreased of about 2 V in few seconds. Successively, the voltage decreased almost linearly up to about 35 Ah discharged, after this value the voltage profile for the three discharging currents rapidly decreased down to the minimum value permitted (10V), when the discharging step finished. The measurement of the energy released by the battery pack during the three discharging tests gave 580, 515 and 505 Wh for discharging at 20, 80 and 100 A, respectively. The ratios between the energy released from the battery during the discharging down to DOD 100% ( $Wh_{OUT}$ ) and the energy fed to it during the charge phase ( $Wh_{IN}$ ) resulted:  $(Wh_{OUT}/Wh_{IN})_{20} = 0.92$ ,  $(Wh_{OUT}/Wh_{IN})_{80} = 0.81$  and  $(Wh_{OUT}/Wh_{IN})_{100} = 0.78$ . This evidenced a clear effect of the discharging current on the possibility to use an unique step to extract from the battery all the energy previously stored, before reaching the minimum safety voltage. This behaviour has to be considered if the storage system is destined to automotive applications, in particular HEV (hybrid electric vehicle) or BEV (battery electric vehicle), where the necessity of continuous and fast acceleration phases can occur. In particular, regarding HEV the above performance could be exploited in those hybrid configurations characterized by a predominant contribution of batteries to dynamic power requirement (plug-in hybrid vehicles or fuel cell vehicles in hard hybrid configuration).

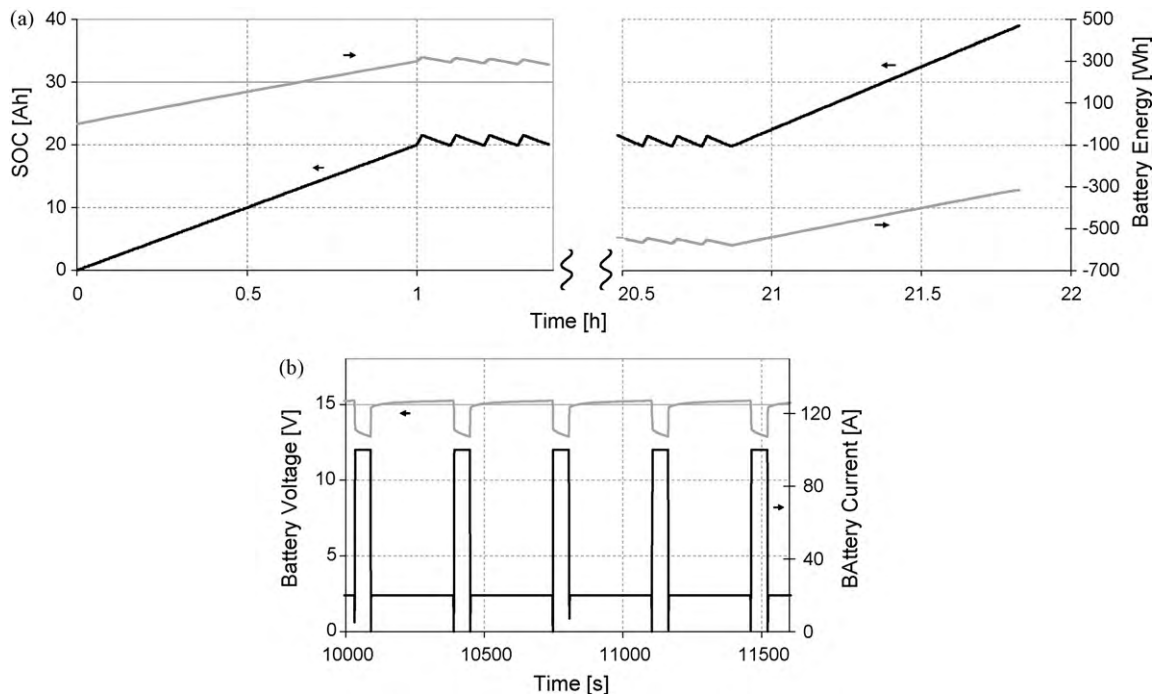
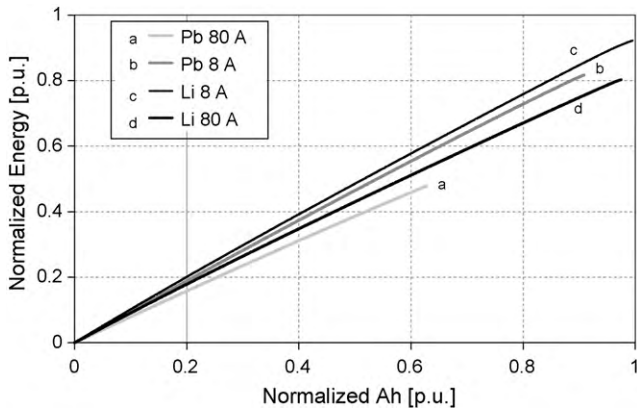


Fig. 4. (a) Characterization test on lithium ion polymer battery pack (12 V, 40 Ah). Discharged Ah and Wh versus cycle length during test cycle 2. (b) Characterization test on lithium ion polymer battery pack (12 V, 40 Ah). Voltage and current versus cycle length during test cycle 2 of (a).

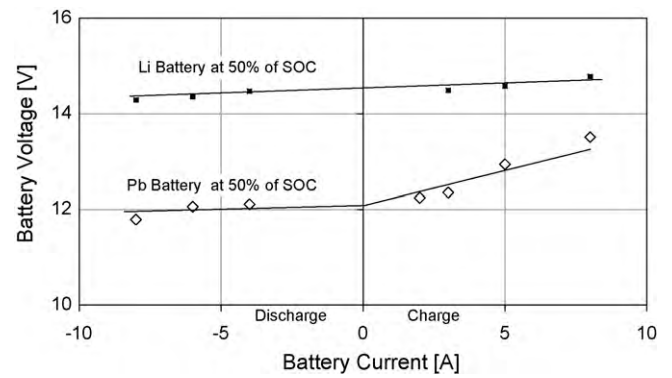


**Fig. 5.** Comparison between lithium ion polymer and lead acid batteries during the discharging phase of type 1 test. Normalized energy with respect to  $Wh_{IN}$  versus normalized Ah at 8 and 80 A.

As when a road vehicle is utilized in crowded urban areas a typical power requirement profile can include many steps of fast load variations (such as a stop-and-go driving cycle), a specific characterization test, described in Experimental as test cycle 2, was carried out on the lithium battery pack. The preliminary charging phase at  $C/2$  has been already shown in Fig. 1, while Fig. 4a shows the results of the discharging period in terms of discharged energy and capacity versus cycle length in hours (for a total of 22 h). Between the initial and final discharging steps at  $C/2$ , a sequence of 200 discharging at  $2.5C$  and charging at  $C/2$  cycles was performed, not reported in Fig. 4a, but zoomed in Fig. 4b in terms of voltage and current as function of time for a period of about 27 min. The total energy released by the battery was 4682 Wh (at 39 Ah), and the ratio of this value to the energy fed during the total charge resulted  $(Wh_{OUT}/Wh_{IN})_{20} = 0.84$ , lower with respect to the value obtained from data of Fig. 1 (0.92), because of the contribution of the 200 discharging steps at 100 A.

In order to appraise the significance of charge–discharge features of lithium batteries, the above results have to be compared with those obtainable with lead acid batteries, which have been adopted on electric vehicles up today. Such a comparison is reported in Fig. 5, where energy values versus discharged Ah are reported in normalized form with respect to  $Wh_{IN}$  of the two batteries, as the nominal capacity of Pb pack was different (38 Ah). The results shown in Fig. 5 refer to discharging phases after type 1 tests effected at 8 and 80 A. The different dynamic behaviour of the two batteries is evident at 80 A, as final ratio  $(Wh_{OUT}/Wh_{IN})_{80}$  resulted 0.81 for lithium and only 0.47 for lead battery, while the DOD 100% condition, when discharge was stopped at safety minimum voltage, was reached at 97% and 63% of SOC for lithium and lead battery, respectively. The better dynamic performance of lithium battery was confirmed also at 8 A, even if the lead system reached a satisfactory value of  $(Wh_{OUT}/Wh_{IN})_8$  (82%) exploiting about 91% of the initial SOC.

On the other hand, an important and specific feature of electric vehicles is the regenerative braking, i.e. the possibility to utilize the energy lost during the conventional braking to charge the battery, that can contribute in a not negligible way to energy economy during road utilization [18]. This feature is obviously favoured when the electric energy storage system is able to accept energy supplied at high power and current. A way to analyze the behaviour of different batteries during charge phases is represented in Fig. 6, where voltage and current values acquired during short time tests of 10–20 s are reported for both lithium and lead acid batteries. The initial SOC of the two batteries was 50%, then they were charged and discharged at constant current in the range 0–8 A. Voltage curves in discharge phases for the two batteries had the same slope, while



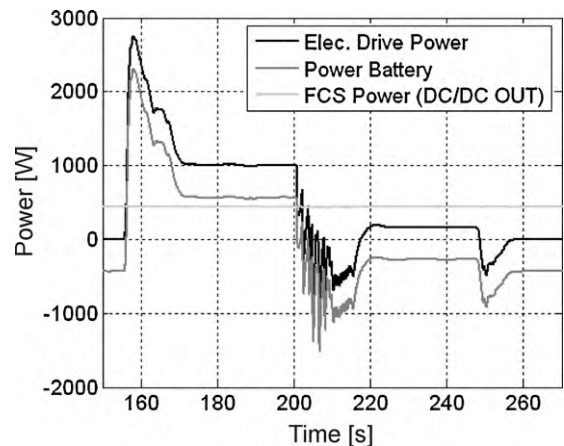
**Fig. 6.** Comparison between lithium ion polymer and lead acid batteries. Battery voltage versus battery current during charge and discharge phases at SOC 50%.

during the charge phases the slope of lead battery significantly increased. The voltage curves reported in Fig. 6 can be represented by the following equation [19]:

$$V = I \times R_{\text{battery}} + V_{\text{OCV}}$$

where  $I$  is the current in Amperes, positive for charging and negative for discharging,  $R_{\text{battery}}$  is the total resistance of the battery and  $V_{\text{OCV}}$  is the open circuit voltage. Then the results of Fig. 6 evidence the higher resistance which causes losses associated to chemical reactions occurring at the electrodes, in particular hydrogen production at anode and oxygen formation at cathode, with water consumption [20]. The behaviour described by Fig. 6 can obviously affect the performance of the storage system when used in a propulsion system on dynamic driving cycles.

The verification of lithium battery performance in real automotive conditions has been effected by using the fuel cell power train designed for scooters and described in Experimental. The test was effected on the R47 driving cycle, which is imposed by European legislation for the evaluation of exhaust emissions of mopeds powered by internal combustion engine, and was used here in the absence of a specific legislation regarding electrical mopeds. The R47 cycle requires the maximum power in the initial phase up to the maximum speed, then a period of constant speed of 50 s at maximum speed, a fast deceleration followed by a second phase of constant speed and a final deceleration up to zero speed. The average power required by the driving cycle resulted 450 W, and this value was controlled at the DC–DC converter outlet in order to realize a hard hybrid configuration.



**Fig. 7.** Characterization of the fuel cell power train for scooter. Power distribution between electric drive, lithium battery and fuel cell system (FCS) during European R47 driving cycle in hard hybrid configuration.

In Fig. 7 the results of a preliminary experiment carried out on the R47 cycle are reported in terms of power distribution between engine, fuel cell and batteries as function of cycle length. The power train test bench was reconfigured by its dedicated software for testing the scooter equipped with the two lithium battery packs, in particular vehicle and road parameters were implemented (weight, rolling resistances and aerodynamic drags).

The engine power reached its maximum value (2750 W) during the first acceleration phase, then decreased down to 1000 W corresponding to the first step at constant engine speed, then diminished down to negative power values during the deceleration phase, when the engine operated as generator. At the end of this deceleration the engine power reached the second condition of constant speed (170 W), followed by the last deceleration up to zero speed. The battery power profile evidences that the storage system compensated the difference between engine power requirements and the power supplied by the fuel cell system. In particular, the engine power peaks were satisfied thanks to the energy coming from the batteries, while during the regenerative braking some energy flowing from the electric machine was recovered into the battery. Furthermore, during the second phase at constant speed batteries were partially recharged by the stack.

Since in the experiment of Fig. 7 the single periods of charge and discharge of the battery pack last not more of 2 min, and current involved was lower than 10 A during the most part of the cycle and under 30 A only during the first acceleration phase, no limitations could be expected regarding battery dynamic performance during the cycle. Having fixed the stack power at the average power required by the cycle, the battery SOC at the end of the cycle was practically equal to that initial. The fuel consumption on the whole driving cycle resulted  $230 \text{ kJ km}^{-1}$ , about 6% lower with respect to that evaluated for the same power train equipped with lead acid battery packs [18]. This decrease of consumption can be mainly attributed to the reduction of vehicle weight (lithium battery packs was about two third lighter than the lead acid packs), and in minor extension to the improvement of energy recovery during regenerative braking due to lithium battery performance.

#### 4. Conclusions

The experimental results presented in this paper permitted to characterize an electric energy storage system based on high power/high density lithium ion polymer cells, in view of an appli-

cation in the automotive field. The lithium systems investigated showed high discharge rate capability, in particular at 2.5C they provided about 92% of their capacity, with a ratio of energy discharged to total energy charged equal to about 80%. This performance resulted significantly better with respect to conventional lead acid systems, for which at about 2C only 62% of the initial capacity was provided. Also in charging mode the performance of lithium batteries resulted more compatible with automotive application, thanks to their minor internal resistance.

The tests carried out on a fuel cell power train for scooter application, equipped with lithium battery packs, on the European R47 driving cycle, confirmed the positive features of these systems observed in steady state characterization, evidencing a satisfactory dynamic behaviour and a fuel consumption reduction of about 6% with respect to the utilization of lead acid systems in the above power train on the same driving cycle.

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