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Abstract

GREENLION is a Large Scale Collaborative Project within the FP7 (GC.NMP.2011-1) leading to the manufacturing of greener and cheaper Li-Ion batteries for electric vehicle applications via the use of water soluble, fluorine-free, high thermally stable binders, which would eliminate the use of VOCs and reduce the cell assembly cost. The project has 6 key objectives: (i) development of new active and inactive battery materials viable for water processes (green chemistry); (ii) development of innovative processes (coating from aqueous slurries) capable of reducing electrode production cost and avoid environmental pollution; (iii) development of new assembly procedures (including laser cutting and high temperature pre-treatment) capable of substantially reduce the time and the cost of cell fabrication; (iv) lighter battery modules with easier disassembly through eco-designed bonding techniques; (v) waste reduction, which, by making use of the water solubility of the binder, allows the extensive recovery of the active and inactive battery materials; and (vi) development of automated process and construction of fully integrated battery module for electric vehicle applications with optimized electrodes, cells, and other ancillaries. Achievements during the first 18 months of the project, especially on materials development and water-based electrode fabrication are reported herein.

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Keywords (separated by '-') Electric vehicles - Energy storage - Batteries - Alloys anodes - Water-based binders - Innovative processing - Battery manufacturing - Automation

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# GREENLION Project: Advanced Manufacturing Processes for Low Cost Greener Li-Ion Batteries

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solubility of the binder, allows the extensive recovery of the active and inactive battery materials; and (vi) development of automated process and construction of fully integrated battery module for electric vehicle applications with optimized electrodes, cells, and other ancillaries. Achievements during the first 18 months of the project, especially on materials development and water-based electrode fabrication are reported herein.

**Keywords** Electric vehicles · Energy storage · Batteries · Alloys anodes · Water-based binders · Innovative processing · Battery manufacturing · Automation

## 1 Introduction and State of the Art

Society's current individual mobility behavior is creating a plethora of looming problems, such as fossil carbon intensity and the concomitant consequences regarding fossil resource supply or the emissions of pollutants such as nitrogen and sulfur oxides ( $\text{NO}_x$ ,  $\text{SO}_2$ ) and particulate matter. While pollutant problems can be addressed by catalytic converters and filters, expectations run high that the greenhouse gas and resource problems can be addressed by substituting internal combustion engine (ICE) cars with battery powered electric cars (BEV). Most of the major car manufacturers have announced BEVs as part of their product lines in the immediate future.

Lithium ion batteries already dominate the consumer portable electronic and telecommunications market due to their higher power and energy density and they are also indicated as the option for the next generation of hybrid and electric vehicles (HEV, EV). The wide deployment of lithium ion batteries in the automotive industry would have tremendous consequences on the battery-market and it would further strengthen the central role of these systems in the field of energy

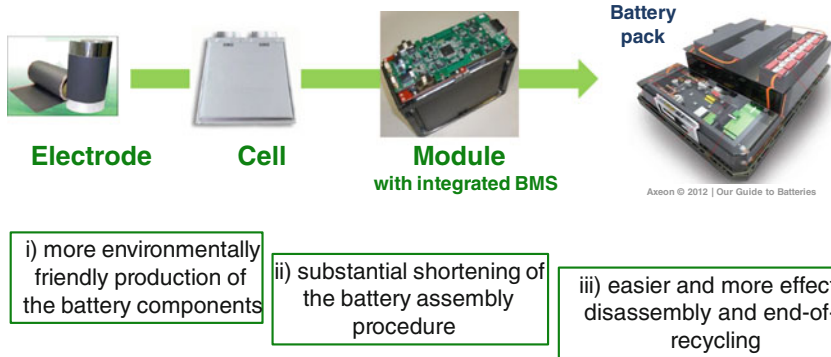
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**Fig. 1** Key levels of battery manufacturing value chain under development in GREENLION

storage. For that, considerable efforts are now focused on the development and realization of lithium ion batteries able to fulfill the requirement necessary for the application in HEV and EV. When the present lithium ion technology is considered, the safety and cost of batteries appear as the main drawbacks holding the introduction of this technology into the automotive market.

In order to tackle these issues from the manufacturing perspective before the final battery pack integration, the GREENLION consortium has identified three key levels in the value chain (Fig. 1), namely battery components, especially electrode processing, individual cells and battery modules, oriented to the battery pack for EVs.

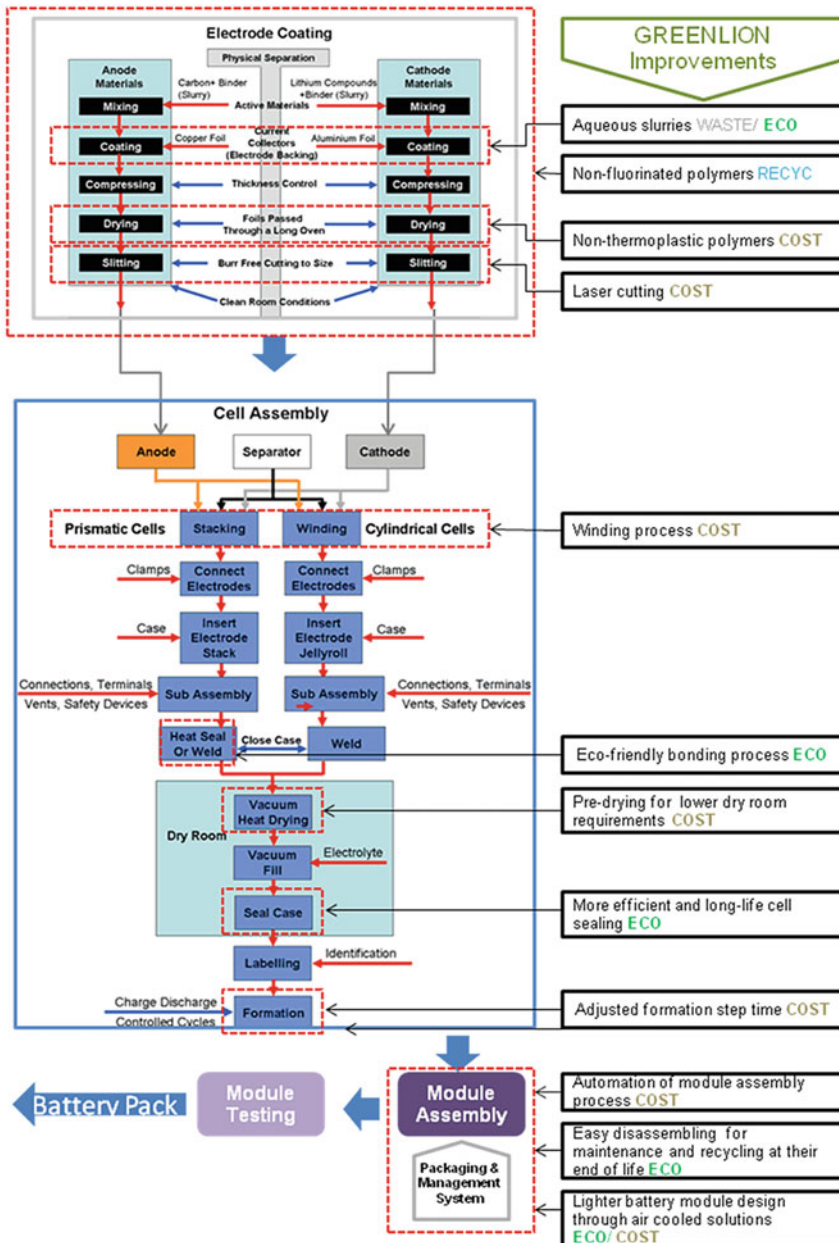
As stated previously, the automotive industry is demanding for safe and low cost lithium ion (Li-ion) batteries to bring to the market higher range and affordable BEVs to substitute the ICE vehicles that won the battle a 100 years ago, due to Ford's mass production line, low oil cost and insufficient battery technology of the time.

The current Li-ion battery manufacturing process has advanced greatly in the last 20 years thanks to the consumer portable electronic industry and those developments are also the basis for the production of large format cells demanded for automotive application. The Li-ion battery production comprises a sequence of steps that can be summarized as electrode preparation, cell fabrication and battery module assembly, as presented in Figs. 1 and 2.

## 1.1 Electrodes

The initial large scale automotive prototype lithium ion batteries use hard carbon or graphite as negative electrode materials, and nickel-substituted cobalt and/or manganese oxide (NCA, NMC), manganese spinel oxide or lithium iron phosphate as cathode active materials. Since for this first stage, the basic formulations and





**ECO:** Eco-friendly; **COST:** Lower cost; **WASTE:** waste reduction; **RECYC:** easier recycling

**Fig. 2** Breakdown of steps for Li-ion battery manufacturing and improvements proposed in GREENLION



73 materials from consumer batteries are being used, the costs of the electrodes still  
74 need to be decreased for the final use in automotive batteries.

75 Numerous groups have been studying and developing new electrode and elec-  
76 trolyte materials with suitable characteristics and improved performance for the  
77 realization of greener and lower cost batteries, and promising results have already  
78 been achieved [1, 2]. However, to realize batteries with such kinds of properties, not  
79 only active and electrolyte materials have to be taken into account but, in general,  
80 all the battery components and even the process to realize the batteries need to be  
81 considered and improved. Apart from the continuous research on materials with  
82 increasing energy and power density, safety, durability and cycle life (targets of at  
83 least 30 % improvement for 2020), current estimations point to a 10–15 %  
84 reduction cost from the active materials that can reach up to 35 % through electrode  
85 process optimization [3].

86 Several studies are now also focused on the improvement of the inactive  
87 materials as well as of the electrode production. In this context, a key role is  
88 certainly played by the binder. As a matter of fact, this component is not only  
89 responsible for the binding of the active materials and the conductive agent to the  
90 metal current collectors, but it also strongly affects the electrode processing.  
91 Consequently, the improvement of the binder must necessarily be considered as a  
92 key point for the development of new safe and greener batteries.

93 An interesting example of the influence of the binder is observed when the  
94 preparation of electrodes based on lithium iron phosphate ( $\text{LiFePO}_4$ ) is considered.  
95  $\text{LiFePO}_4$  displays high stability of the capacity during prolonged cycling; it is  
96 environment-friendly, cheap, and safe [4, 5]. Because of these characteristics, it is  
97 considered as a very attractive cathodic material. So far, however, most of the  
98 research and the development in composite cathodes have been focused on the use  
99 of fluorinated binders and practically all commercial lithium-ion batteries are made  
100 using poly(vinylidene fluoride) (PVDF) as the binder. However, this polymer is  
101 costly (industrial cost in the multiton scale is around 15–18 EUR/kg); it requires the  
102 use of volatile organic compounds that are often toxic (like N-methyl pyrrolidone,  
103 NMP) in the processing, and it is not easily disposable at the end of the battery life.  
104 The introduction of alternative binders, as well as an improved preparation pro-  
105 cedure, is necessary.

106 Recently, alternative binders have been introduced for the manufacture of  
107 anodes for lithium-ion batteries, like styrene butadiene rubber (SBR) that can be  
108 processed in water. Among them, one of the most interesting is certainly the sodium  
109 salt of carboxymethyl cellulose (CMC), which is a water-soluble material. This is  
110 certainly the greatest advantage of CMC because it allows processing in aqueous  
111 slurries rather than in polluting, health and environment unfriendly, volatile  
112 organic-compound-based slurries. The second great advantage of CMC resides in  
113 its easy disposability at the end of the life of the battery. Once the electrode is  
114 extracted, the active electrode material can be easily recovered by pyrolysis of the  
115 binder. Last but not least is the material cost. The CMC industrial price is about 1–2  
116 EUR/kg, i.e., about 1 order of magnitude lower than PVDF.



## 1.2 Cell Assembly

Battery pricing is significantly impacted by material costs and manufacturing cost in mass production due to the multitude of operations and the precision required. The electrode thickness produced at the first step can range from 0.05 to 0.2 mm depending on the electrode type (cathode or anode), the intended application of the battery (high capacity or high power) and the cell design (cylindrical or planar).

Cylindrical cells, where components are staked and wound to be inserted into cylindrical cases, and prismatic cells, with stacked electrodes and separators, are currently the predominant designs. Pouch cells are prismatic cells with aluminum-polymer soft pack instead of metal can, so they achieve a packaging efficiency of 90–95 % and higher energy density. With high volume, any reasonable size can be produced economically. Lithium polymer pouch cells are increasingly being considered as alternatives to large prismatic cells for automotive applications; because their form is flexible they can be packaged more efficiently, and reduced cell packaging overheads result in high battery energy density. Due to large surface area and aspect ratio they have good heat dissipation. However, the cells have low mechanical stability and therefore more robust packaging is required.

## 1.3 Module Design and Assembly

In order to develop a battery module as a building block of a battery pack, first of all it is highly convenient to have as much information as possible about the characteristics of the vehicle to be powered by the energy stored in the battery in terms of weight, friction, aerodynamic coefficient, efficiency, voltage and current of the power train, ... Besides, others features related to the vehicle performance must be defined, as the energy storage will be sized in order to cope with these requirements, such as autonomy, acceleration, maximum speed, cruise speed, etc., referred to a given driving cycle.

According to the vehicle characteristics and requirements, and once the cell has been selected, tested and modeled, all this information will be used to determine the required number of cells and modules and their series/parallel connection inside—respectively—the module and the battery pack, so that the required voltage, current, energy and power values are met. In order to define the optimum possible arrangement, the resulting module will be simulated—both electrical and thermally—out of the previously obtained models of the cells. Special consideration should be put in the lay-out of the cells since the thermal behavior of the module will strongly depend on this.

Most of the systems using batteries require a certain number of cells connected in series and parallel in order to achieve the desired voltages and current. Therefore, all the cells should be kept in the same state of charge (SOC) in such a way that the capacity of the resulting module or battery-pack is not reduced due to a weak cell



156 that reaches the cut-off voltage sooner than the rest, or to an incomplete charge  
157 caused by a cell with a voltage higher than the others. Therefore, a cell balancing  
158 system that keeps the cells in the same SOC is required to improve the performance  
159 of the module.

160 Besides, special care must be taken in order to ensure that no cell is over charged  
161 or discharged, due to the electrochemical inequalities of the cells inherent to the  
162 manufacturing process or to uneven working or balancing conditions. Otherwise, the  
163 users' and cells' integrity could be compromised, as dangerous amounts of flam-  
164 mable gases and/or toxic chemicals can be released, and even end up in an explosion.

165 In order to have an optimal use of the module, it is highly convenient to have  
166 access to the information concerning the state of charge (SOC, which is the  
167 remaining charge in the cells) and state of health (SOH, which is the capacity of the  
168 cells at a given time compared to that when they were new) of the cells. For all this  
169 reasons, a Battery Management System (BMS) is required in any system using  
170 lithium-ion cells.

171 The operation of batteries depends on an electrochemical process for both  
172 charging and discharging, and it is widely known that these chemical reactions are  
173 significantly dependent on temperature. Nominal battery performance is usually  
174 specified for working temperatures somewhere in the +20 to +30 °C range. How-  
175 ever, the working temperature conditions of the cells can deviate substantially from  
176 nominal values, in such a way that batteries are operated at higher or lower tem-  
177 peratures. As a consequence, the performance of the cells is strongly affected: in  
178 general terms, discharge time (and therefore, capacity) decreases at lower temper-  
179 atures, and the number of charge and discharge cycles is reduced when working at  
180 higher temperatures.

181 Besides, and from a safety point of view, it is extremely important to avoid a  
182 thermal runaway (uncontrolled temperature increase) in the cells since dangerous  
183 amounts of flammable gases and/or toxic chemicals can be released, and even end  
184 up in an explosion.

185 Therefore, a Thermal Management Systems (TMS) is required to maintain the  
186 cells within a safe temperature range that, besides, allows optimizing the perfor-  
187 mance of the module. In order to cool-down or heat-up the cells, different systems  
188 can be used, being air or liquid cooling the most usual choices.

## 189 2 Project Description

### 190 2.1 Project Approach and Objectives

191 In the GREENLION project ([www.greenlionproject.eu](http://www.greenlionproject.eu)), we address the issues cited  
192 previously by the industrial development of eco-designed processes at the electrode,  
193 cell and battery module level. At the **electrode processing stage** (that will be  
194 otherwise independent of the active materials chemistry), developing and making  
195 use of:

- 196 1. aqueous slurries rather than toxic organic volatile compounds (25 % cost  
197 reduction);
- 198 2. non-thermoplastic polymers that allow for high temperature drying, which  
199 results in shorter and less expensive assembly procedure (10 % efficiency); and
- 200 3. easily disposable non-fluorinated polymers (at expected 10 times less materials  
201 cost).

202 At the **cell assembly level**, further improvements to the existing procedures as  
203 well as changes at some steps of the assembly process will be developed to increase  
204 energy efficiency and shorten times (and hence lower costs) during the manufactur-  
205 ing process, by implementing:

- 206 1. laser cutting instead of mechanical notching of the electrodes (15 % cost),
- 207 2. adjusted stack winding of components from aqueous-based electrodes and their  
208 drying process before electrolyte filling and sealing, to lower dry room  
209 requirements,
- 210 3. environmentally friendly bonding process for more effective and long-life cell  
211 sealing, and
- 212 4. adjusted formation step time (ideally for electrodes with reduced formation  
213 cycle) in cell manufacturing line (5 % time reduction).

214 Finally, developing a modular battery allows an easier handling of cells within a  
215 complete battery pack. At this **battery module level**, GREENLION project will  
216 design an autonomous unit including its own electrical and thermal management as  
217 a simple and reliable building block that will allow the manufacturing and main-  
218 tenance of the whole battery packs easier and more inexpensively, with the lowest  
219 possible environmental impact. This will be achieved by:

- 220 1. lighter battery module designs (including electronics) with the possibility of  
221 implementing air cooled solutions instead of liquid cooling systems (expected  
222 20 % less weight),
- 223 2. bonding process of module housing for safe operation but easy disassembling  
224 for maintenance and reuse/recycling at their end-of-life, and
- 225 3. automation of module assembly process (3 s/cell vs. manual assembly).

226 These developments will be scaled-up and realized in pilot lines during the  
227 project, following a continuous environmental assessment of materials and pro-  
228 cesses. A validation of the finally assembled battery module will be carried out lead  
229 by the automotive end-user who will also provide the targets and specifications for  
230 (H)EV application.

231 General project approach and objectives are summarized in Fig. 2. Progress  
232 beyond current State of the Art is also indicated.

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## 2.2 Project Consortium

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The scientific and technological cooperation in GREENLION consortium and their roles in the project are well balanced covering the complete chain from raw material, scientific comprehension, technological research and end users. As an essential part of the project, the industrial partners will commit to exploit all commercial aspects of the new manufacturing processes. To this purpose, the industrial partnership was designed to combine Li-ion cells and module manufacturers (CEGASA), processing equipment manufacturers (POLYTYPE and KEMET), material suppliers (SOLVAY and TIMCAL), automation of assembly processes providers (MONDRAGON ASSEMBLY), recycling and waste treatment services suppliers (TECNICAS REUNIDAS), and car manufacturers (SEAT and VOLKSWAGEN). The research institutions (CIDETEC, ENEA, RESCOLL, AIT) and universities (University of Muenster, Politecnico di Milano, University of Limerick) in GREENLION consortium provide complementary skills and expertise in the relevant fields of research and development that are necessary to achieve the project objectives.

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## 3 Outcome of the Project

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### 3.1 Baseline for GREENLION Project and Performance Indicators

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Knowledge will be generated well beyond state of the art and the limitations of current Li-ion battery manufacturing process. In particular, Table 1 summarizes and quantifies the most significant targets.

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### 3.2 Expected Impact of the Project

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GREENLION will provide advances to a number of scientific and engineering challenges for battery cell and module manufacturing, and their performance thereof. The successful resolution of these will lead to breakthroughs in automotive lithium ion batteries for electric vehicles and thus to the development of a sustainable mobility and quality of life.

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Greening our transport system is necessary not only to avoid the influence of oil supply (\$147 per barrel peak in 2008) but also to achieve EU and international targets in emissions reductions. In the EU, 19 % of total greenhouse gas emissions and 28 % of CO<sub>2</sub> emissions in 2005 are linked to the transport sector. More than 90 % of the total EU transport-related emissions are due to road transport. While



**Table 1** Summary of most relevant GREENLION performance indicators and targets

	Proposed innovation-performance indicators
Electrode	<p>Development of innovative electrodes realized by water-based processes to realize electrodes characterized by: (i) high thermal stability to allow high-temperature drying (&gt;150 °C) in order to allow assembly in less stringent dry room operating conditions and reduce post-coating treatment time); (ii) high electrochemical stability to allow the use of high voltage cathodes (at least 5 V vs. Li/Li<sup>+</sup>); (iii) high capacity retention upon cycling (more than 80 % of initial capacity after 1,000 cycles)</p> <p>The final goal is to obtain anodes and cathodes for lithium-ion batteries with storage capacities as high as, respectively, 300 and 150 mAh/g (excluding the weight of the current collectors), and surface loadings of, at least, 5 mAh/cm<sup>2</sup></p>
Cell	<p>Implement laser cutting/slitting instead of mechanical notching of the electrodes achieving negligible degradation of active material in the cut area, reduction of burrs resulting in a safer cell, expected 15 % cost saving due to reduced maintenance and higher process efficiency</p> <p>Based on such innovative electrodes, GREENLION proposes to design and develop cells capable of delivering a specific energy of 200 Wh/kg, which is the actual target for automotive applications</p>
Module	<p>Lighter battery module designs (including electronics) by evaluating the implementation air cooled solutions instead of liquid cooling systems (20 % less weight)</p> <p>Automation of module assembly process with a handling time down to 3 s per cell) will enable cost reduction and quality in line with what achieved in the highly automated cell manufacturing</p>

266 total EU emissions declined, transport emissions increased continuously between  
 267 1990 and 2005 due to high growth in both passenger (28 %) and freight transport  
 268 (62 %).

269 Current and near-term (i.e. Li-ion) battery technology development is one of the  
 270 key factors on the Mobility Electrification and the large scale production of these  
 271 automotive batteries and reducing their costs is, in fact, critical for market entry and  
 272 acceptance of Electric Vehicles. In order to achieve a break-even cost with internal  
 273 combustion engines, battery costs must be reduced from the current estimated range  
 274 of 675–500 € per kilowatt-hour (kWh) at high volume production (order of 100 k  
 275 units) down to 350–275 €/kWh by 2020. R&D to improve power (W/kg) and  
 276 energy density (Wh/kg) in order to increase driving autonomy, reductions in  
 277 recharge time and achieving life cycles that approach vehicle life spans is also  
 278 imperative. Increasing production rate from 10,000 to 100,000 batteries/year  
 279 reduces cost by ~30–40 % [6].

280 GREENLION addresses further reduction costs driven not only by high volume  
 281 manufacturing, but also from the components processing conditions. The use of water  
 282 based binders, an order of magnitude cheaper than conventional fluorinated ones will  
 283 drive down the cell manufacturing costs, besides being more environmentally  
 284 friendly and eco-sustainable at the end of life of the cells. Besides the improvement in  
 285 environmental, health and safety terms (including “working-condition-friendly”

considerations), the initial inversion and running costs of the solvent recovery system would be avoided and water is indeed cheaper than NMP. Even though in current production plants the recovered NMP is purified and offered again at 50 % of the cost of pure solvent, distilled water is also cheaper (0.20 €/L) than 50 % of pure NMP 0.90 €/L (~ 1.8 €/L pure).

Expected impact in the field of new competitive processes, by means of production automation is also foreseen. Not only will the results of the project efforts enable lower cost and greener lithium battery packs production, but also equipment manufacturing and high added value processes will be developed. These new automated processes will contribute to a substantial cost reduction of lithium battery packs, and will facilitate their introduction to mass production.

Automation and new process development will improve the quality and yield of the production, while at same time reduces labor costs per kWh. This project, with the development of the specific equipments for module assembling, will enable a cycle time of 3 s for each cell. This results in a module production capacity of 880 MWh/year.

Globally, automation and equipment development in this project will enable a cost reduction of the whole battery pack of 15 %. Having in mind that only 24 % of the cost is related to the module/pack manufacturing (60 % are materials components and 16 % are transports and others), it represents a major step in the way to mass production. Market growing will also pull down the prices of the materials, and it will open the way to the mass production at competitive costs.

### 3.3 Results Achieved

GREENLION is currently at month 18 of a 4 year-long workplan. During this first stage of the project, efforts have been mainly focused on the electrode processing step, with the development and testing of active materials and binders suitable for water-based slurry formulations and electrode coating process. First selected formulations have been used for small scale GEN1 prototype pouch cell assembly while the optimized module design and assembly process is underway. These results and advances are summarized in the following sections.

#### 3.3.1 Materials Development and Water-Based Electrode Processing

Among the main research topics of the GREENLION Project, are to be highlighted the development of ionic liquid-based electrolytes and the realization of electrodes, prepared through innovative, eco-friendly process routes, based on high-voltage cathode and large-capacity anode materials. There is growing up interest in replacing the organic solvents currently used in lithium batteries [7–9] with ionic liquids, ILs, since their non-flammability and negligible vapor pressure in conjunction with wide chemical, electrochemical and thermal stability, high ionic



conductivity and heat capacity. Our basic idea is to favorably combine different IL sets in order to obtain ionic liquid mixtures with improved performance. For instance, N-methyl-N-propylpyrrolidinium bis(fluorosulfonyl)imide (PYR<sub>13</sub>FSI) was found to exhibit moderate viscosity and low melting point, allowing fast ion conduction even at low temperatures [10]. On other hand, the much cheaper N-methyl-N-propylpyrrolidinium bis(trifluoromethanesulfonyl)imide (PYR<sub>13</sub>TFSI) shows wider thermal and electrochemical stability [11]. In order to verify if these characteristics could be combined, PYR<sub>13</sub>FSI-PYR<sub>13</sub>TFSI mixtures were prepared and investigated in terms of NMR spectroscopy, transport properties and density measurements.

Remarkable conduction values, e.g., approaching  $10^{-3} \text{ Scm}^{-1}$ , are achieved already at  $-20^\circ \text{C}$  for mole fraction ranging from  $0.6 \leq x \leq 0.8$  whereas both the raw ionic liquid materials (PYR<sub>13</sub>FSI and PYR<sub>13</sub>TFSI) are solid at this temperature (see Fig. 3). This highlights the synergic effect exhibited in ionic liquid mixtures, especially for low temperature applications.

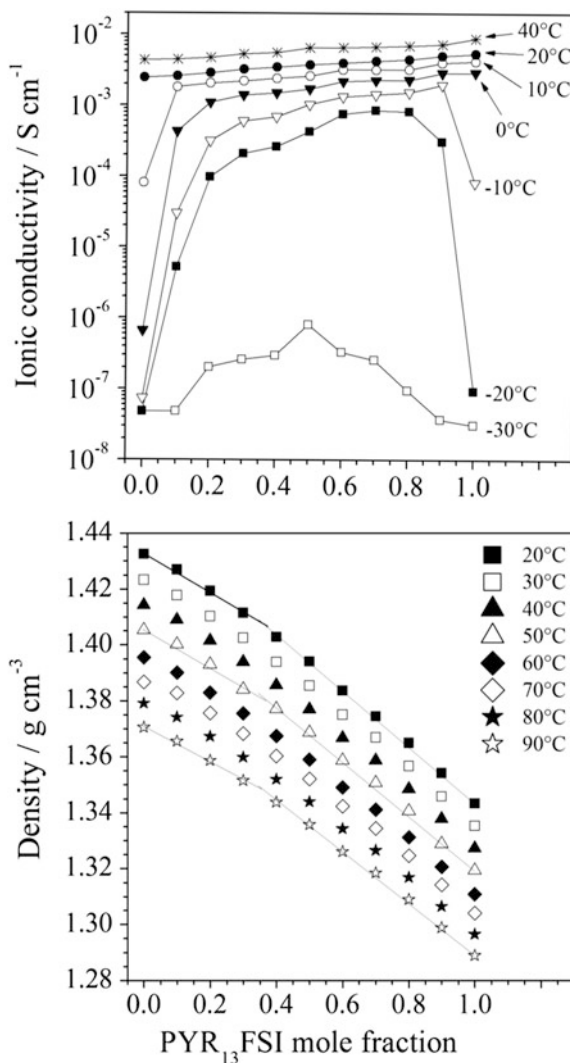
NMR heteronuclear NOE correlation experiments (HOESY) experiments have been successfully used for the assessment of the intermolecular contacts between the F atoms of the anions and the H atoms of the cations in pyrrolidinium based ionic liquids, thus providing information on the local structural organization. The experiments have shown a peculiar cation-anion organization in the three investigated blends responsible of their favorable physico-chemical characteristics.

High nominal voltage cathode materials, combined with large capacity anodes are appealing issues for the realization of lithium batteries with high gravimetric and volumetric energy. In this first stage of GREENLION, cathodes based on LiNi<sub>x</sub>Mn<sub>y</sub>Co<sub>1-x-y</sub>O<sub>2</sub> (NMC) and anodes based on carbonaceous materials (graphite, SLP) have been developed. The composite electrodes were fabricated using the fluorine-free, water-soluble, natural binder carboxymethylcellulose sodium salt (CMC) instead of the more expensive and less environmentally friendly polyvinyliden-di-fluoride (PVdF) in N-methyl-pyrrolidone (NMP). The use of CMC allows also easier recycling of the battery components. For instance, the dissolution in water of the binder allows, for example, a full recovery of the metallic current collectors [12, 13].

Commercially available NMC cathode and Timcal SLP 30® graphite anode tapes based on the aqueous CMC binders were prepared using a pre-pilot automated coating line. The cycling performance tests (Fig. 4) evidenced a time-stable capacity of  $130 \text{ mA h g}^{-1}$  for more than 40 cycles with coulombic efficiency higher than 99.0 % for the NMC cathodes. The SLP 30® anodes showed very good performance in terms of reversibility of the intercalation process. The specific capacity leveled  $375 \text{ mA h g}^{-1}$  after a few cycles. Upon 80 cycles, the SLP 30® electrodes showed still high cycling stability and coulombic efficiency above 99.9 %. These results support for a further development of the aqueous CMC binder-based electrodes.

In addition, alternative water-soluble binders have been studied. Impressive electrochemical performance has recently been reported for Si nanopowder [14] and nanowire [15] anodes prepared from aqueous slurries using 15 wt% alginate as

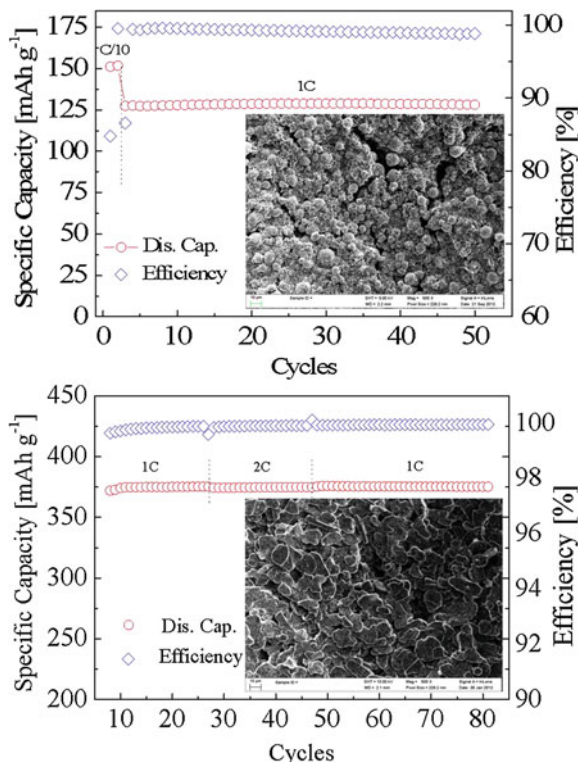
**Fig. 3** Ionic conductivity (*upper panel*) and density (*lower panel*) versus  $\text{PYR}_{13}\text{FSI}$  mole ratio dependence for (x) $\text{PYR}_{13}\text{FSI}/(1-x)\text{PYR}_{13}\text{TFSI}$  binary electrolyte mixtures at different temperatures



369 binder. To date, however, no study has been undertaken using alginates as the  
 370 binder for graphitic anodes, which are almost ubiquitous in present lithium ion  
 371 battery technology. It is apparent from Fig. 5 that the graphite anode with 7.5 wt%  
 372 alginate outperforms that with 10 wt% PVDF (a common commercial level) over  
 373 the course of the first 65 charge/discharge cycles. This result suggests that alginate  
 374 may be a suitable candidate for aqueous manufacturing of anodes.

375 The CMC based formulations will be the first to be trialed in pilot line in order to  
 376 develop optimized coating machinery and electrodes for cell assembly. The most  
 377 efficient way of manufacturing battery electrodes is to simultaneously coat both

**Fig. 4** Cycling performance versus Lithium and SEM image (*inset*) of NMC cathode (*upper panel*) and SLP 30® anode (*lower panel*) in (1 M)  $\text{LiPF}_6/\text{EC}:\text{DMC}$  (1:1 in wt/wt) electrolyte at 20 °C. Current rate: 0.1–1 C; mass loading: NMC,  $3.29 \text{ mg cm}^{-2}$ ; SLP 30®,  $2.45 \text{ mg cm}^{-2}$



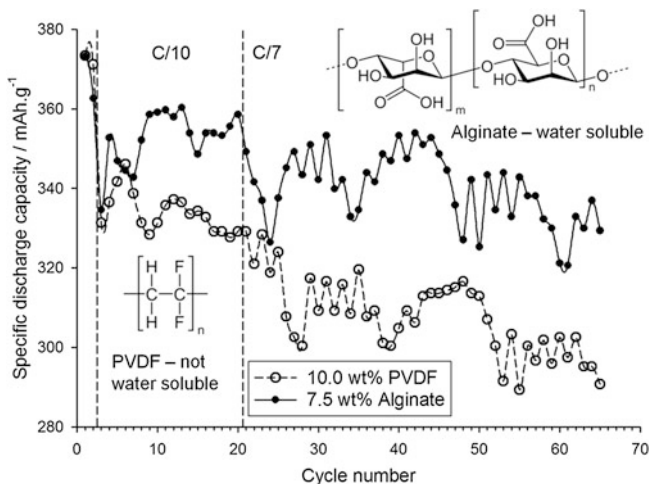
378 sides of the substrate and to use a flotation dryer for removing the solvent. This  
 379 configuration requires one of the coatings to be applied in the so called kiss coating  
 380 mode as depicted in Fig. 6 for the slot coating process.

381 Coating trials have been carried out on a pilot machine, allowing the adjustment  
 382 of parameters to achieve an excellent uniformity of the kiss-coated layer, i.e. by  
 383 suppressing cross lines generated by web flutter in the flotation dryer, and by sup-  
 384 pressing longitudinal bands generated by web deformations upstream of the slot die.

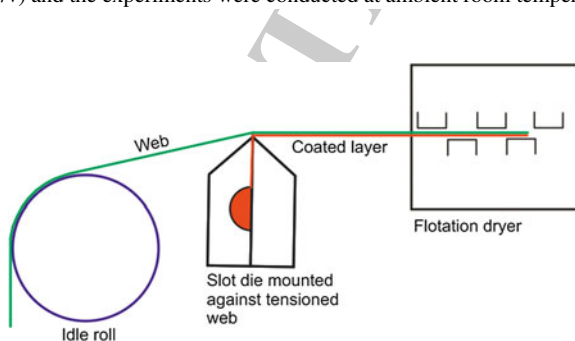
### 385 3.3.2 Cell Assembly and Module Design

386 During the first year, GEN0 prototype cells (10–14 Ah) were assembled as baseline  
 387 for the project, from electrodes prepared with commercially available water-soluble  
 388 binders and graphite/ $\text{LiFePO}_4$  (C/LFP) chemistry.

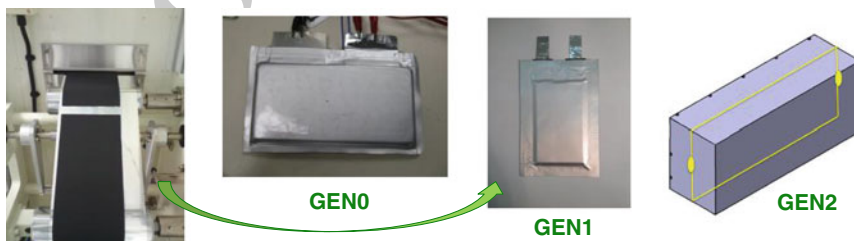
389 The NMC and SLP 30® electrodes (around  $1 \text{ m}^2$ ) prepared in a pre-pilot  
 390 automated coating line were used to assemble GEN1 small pouch cells  
 391 (0.5–1.5 Ah) as shown in Fig. 7, following the first large cell design (30 Ah target)  
 392 proposed to fulfill the energy requirements of the end-users for an efficient auto-  
 393 motive battery module.



**Fig. 5** Comparison of the specific discharge (delithiation) capacities of anodes prepared using polyvinylidene fluoride (PVDF) or alginate binders. The only other component of the anodes was the active graphite material—TIMREX® SLP30 by TIMCAL. The first and second charge/discharge cycles were conducted at slow rates of C/40 and C/25 respectively to facilitate the formation of a stable SEI layer. Voltage limits were between 5 mV and 1.5 V versus Li/Li<sup>+</sup>. The electrolyte was 1 M (EC:DMC, 1:1 v/v) and the experiments were conducted at ambient room temperature



**Fig. 6** Schematic view of coating the web-underside by the slot coating process operating in the kiss or tensioned-web mode



**Fig. 7** From left to right CMC-based electrodes prepared in pre-pilot coating line, *GEN0* C/LFP cell, *GEN1* small pouch cell with SLP 30® and NMC electrodes and schematic *GEN2* power cell and module design

394 While laser notching trials of electrodes with both PVDF and water-based  
 395 binders are underway, conventional cutting dies (mechanical notching) and manual  
 396 stacking process were used for GEN0 and GEN1 cell assembly. Automated  
 397 stacking-winding will be implemented for the GEN2 cell that has been adopted as  
 398 the most efficient electrical and thermal design for high power performance.

399 The design of a lighter battery module suitable for automated assembly and  
 400 easier disassembly is ongoing, coupled to the GEN2 power oriented cell design.  
 401 Different aspects such as minimum mechanical fitting by the assembly process,  
 402 modular assembly including liquid cooled cold plates, mechanical absorption of cell  
 403 swelling and venting are under consideration.

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