Lithium-Air Batteries with split Oxygen Harvesting and Redox processes

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LABOHR

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Final Report Summary - LABOHR (Lithium-Air Batteries with split Oxygen Harvesting and Redox processes)

Executive Summary:
The LABOHR project, ‘‘Lithium-Air Battery with Split Oxygen Harvesting and Redox Processes’’ was a collaborative project funded by the European Commission under the FP7-2010-GC-ELECTROCHEMICAL-STORAGE and coordinated by Stefano Passerini from Wesfälische Wilhelms-Universität Münster (WWU).
LABOHR has put together a consortium, with complementary competencies in the fields of material synthesis and electrochemistry from both academic partners [Wesfälische Wilhelms-Universität Münster (WWU), Tel Aviv University (TAU), Consejo Superior de Investigaciones Cientificas (CSIC), University of Bolgna (UNIBO), University of Southampton (SOTON) and Kiev National University of Technology and Design (KNUTD)] and industrial partners [SAES getters S. p. A. (SAES) and Rockwood Lithium (ROLi)], while the engineering and specifications were relying on industrial partners [AVL List GmbH (AVL) and Volkwagen AG (VW)]. In addition, a management company [European Research Services (ERS] was involved for ensuring good communication and helping the coordinator with project management.

LABOHR aimed at developing a Li-air battery working prototype operating in flooded configuration and using ionic liquid-based electrolytes. The specificity of the project was the use of the electrolyte as an oxygen carrier to harvest O2 from ambient air through an external O2 harvesting device, so that a O2 saturated electrolyte is circulated via flowfield plates instead of O2, with the goal of solving the cathode clogging problem, by a more homogeneous deposit of the reaction products within the porosity of the cathode, operating in flooded configuration.

To achieve this target, all the key issues of the cell chemistry have been addressed: The anode (lithium or silicon/carbon nanocomposite electrodes) as well as the stabilization of its interface with the electrolyte, the cathode (porous carbon, functionalized or not with catalysts), the electrolyte (ionic liquid-based) and the oxygen harvesting. In the meantime, more fundamental studies were performed for the purpose of a better understanding of the different aspects and limitations of the technology, while feeding the modeling tasks with the required physico-chemical parameters

Beside the chemical aspects, a full Li-air pack has been designed and modeled, to assess the ability of the technology to meet the specifications of the automotive industry, as well as to define realistic target values for all the technical aspects. Finally, although the technology is still at the research level, eco-design, sustainability and recycling have been considered as well.

Project Context and Objectives:
Li-ion batteries are nowadays the best secondary battery technology due to their high energy density and, after having been used for more two decades in low power portable applications such as camcorders, cameras, cellphones and laptops, are now conquering a new market: Electric vehicles. However, most electric cars only allow for a driving range slightly above 100 km in the most favorable driving conditions, which corresponds to the reserve of most gasoline cars and, if the Tesla model S allows for a 400 km range, it includes a 85 kWh Li-ion battery pack which, alone, costs twice the price of a standard economy car making it a niche market reserved to the happy few that can afford it. On the other hand, most electric cars include smaller Li-ion packs and, although being more expensive than their gasoline equivalents, have a range that prevent their use for anything else than a commuter (second) car. This is due to the intrinsic limitation of insertion electrodes on which is based the Li-ion technology and, if progresses are still foreseen, by increasing the capacities of the electrodes or the operating voltage of batteries, a practical limit is expected to be reached within the next decade as a result of the weight of the carbonaceous anodes and the heavy transition metal-based cathodes, needed for the insertion of Li+, which results in an anticipated limit of energy density between 200 and 250 Wh kg-1 , depending on the authors.

On the contrary, the Li-air battery would operate with a Li metal as anode, which sports a specific capacity of 3860 mAh g-1, to be compared with the 372 mAh g-1 of graphite, currently used in Li-ion anodes. The cathode is even more promising, given that O2 is virtually a ‘no cost’ cathode harvested from air and would, in principle, be stored on board only in the discharged state (under the form of Li2O2) which results in an theoretical energy density in excess of 3000 Wh kg-1, postulating a 2.7 V discharge voltage. Although a cathode porous substrate is, in fact, needed for hosting Li2O2, considering 3000 mAh g-1 for the cathode substrate still leads to calculated energy densities above 2000 Wh kg-1, that is a ten folds increase as compared with the best foreseen Li-ion batteries.

Although the technology is not mature yet, especially compared to Li-ion, it justifies intense research by many groups around the world, constantly revealing new issues and proposing new ideas and concepts to overcome them, comforting Li-air as the ‘holy grail’ of secondary batteries.

One of the issue, rarely highlighted, is the fact that, if the air (or O2) cathode is very similar to that of a fuel-cell, the main difference is that oxygen reduction leads, after chemical reaction with Li+, to the deposition of solid Li2O2 within the porosity of the cathode substrate. In addition, Li metal being not stable versus aqueous electrolytes, organic electrolytes must be used. Moreover, the operation of fuel-cells is linked to the maintaining, by use of perfluorinated hydrophobic polymers, of 3-phases contact points within the cathode (that are contacts between carbon, ensuring electronic transport, aqueous electrolyte, ensuring H+ transport-in the case of PEMFC-, and O2), which allow for a fast feed of O2 and fast oxygen reduction reaction. The discharge product, water, being evacuated in the atmosphere. In the case of Li-air, on the contrary, the porosity as well as the hydrophobicity of the cathode changes with discharge, making questionable its steady operation in a 3-phases configuration. Several authors also pointed out that operating Li-air cathodes in a flooded configuration, (i.e. a 2-phase configuration relying on dissolved O2 only), would allow for more homogeneous deposition of Li2O2 and would lead to less porosity clogging, which is one of the reasons for capacity limitation as it prevents O2 access to some areas of the cathode. As a matter of fact, most results are obtained using very thin electrodes in the 1 mg cm-2 range, operating at the interface between (a closed) O2 atmosphere and the electrolyte, for minimizing the diffusion distance for O2, a model that cannot be extrapolated to the high capacity batteries of electric vehicles , given the high O2 flow that would need to be provided to the Li-air pack.

LABOHR approach was radically different, as it proposed to operate Li-air battery in flooded configuration, the O2 saturated electrolyte being circulated from an external O2 harvesting device to the cathode. With this concept, forced convection (i.e. electrolyte flow) allows minimizing the diffusion distance for O2, one of the limiting parameters for reaching high capacity and power. It also avoids having to blow O2 directly in the cathode, an environmentally questionable approach, given that any electrolyte leaking out or evaporating from the cell would result in atmospheric pollution, the electrolytes used being organic. To minimize even further any risk of electrolyte loss, the use of ionic liquid-based electrolytes, which are totally non-volatile was proposed. From a safety point of view, in batteries incorporating Li metal (which reacts violently in presence of humid atmosphere), their non-volatility, associated to their non-flammability and high temperature stability is definitely a plus. Their hydrophobicity and good compatibility versus the Li metal anode complete their competitive advantages, in addition to that fact that the oxygen reaction products have been found to react with most solvents within the duration of LABOHR, confirming he validity of their use.

The approach of splitting the O2 harvesting form the electrochemical reactions taking place at the anode and cathode had another goal, considering a technology in which both anode and cathode operation need thorough investigation and development: Avoiding the influence of the O2 direct feed, which is highly dependent on the operation conditions. In addition, as the final practical target of Li-air battery, linked to their high theoretical energy density, is the powering of electric vehicles, the technology, including the O2 harvesting from air and the circulation of the electrolyte had to be evaluated in regard of the specification of the automotive industry and the full car powering system, including ancillaries had to be designed and modelled, for accessing the target values for laboratory experiments. As a candidate for future mass market electrical vehicles, eco-design and sustainability had to be evaluated as well.

Thus, the objectives of LABOHR, can be summarized as follow per (technical) work package (WP):

WP3: Definition of the specification and the cell chemistry from the need of the car industry, led by Volkswagen AG

WP4: Design and modelling of Li-air pack and cells, including all the ancillaries needed for its operation, led by AVL list

WP5: Development of high capacity anodes both made of Li metal and/or Si-carbon nano-composites and the stabilization of their interface with the electrolyte, led by University of Tel Aviv

WP6: Study of all the key issues related to cathode operation (led by University of Bologna):
o Determination of all the parameters linked to the oxygen redox reaction (diffusion coefficients and

solubility of O2 in ionic liquid, as well as kinetic constants)
o Development of carbon-based porous cathode by use of template methods
o Preparation and evaluation of non-noble catalysts
o Determination of the nature of reaction products

WP7: Development of an oxygen harvesting device, able to harvest O2 from air to feed a Li-air battery, led by SAES getters

WP8: Development of a lab-scale prototype including an O2 saturated ionic liquid-based electrolyte circulation and a flowfield plate, led by Westfälische Wilhelms-Universität Muenster.

Project Results:
The advances brought by LABOHR to the Li-O2 field can be witnessed by the amount of scientific publications which have been published during the course of the project (27 so far), in addition to three patent applications by AVL concerning different aspects of Li-air pack use for automotive application.

As one of the primary objective of WP6, the O2 redox reaction was studied in pure ILs as well as in Li salt containing electrolytes, different electrochemical methods (Rotating disk electrode (RDE), ultra micro electrode (UME), voltamperometry), in some cases combined with modeling, were implemented, the results of these studies, which allowed a better understanding of the fundamental factors limiting the performance of Li-air batteries were reported within the following articles by UNIBO (with a collaboration with WWU) and SOTON, who also studied the O2 electrode reaction products, introducing a new in-situ Raman cell:

1- F. De Giorgio, F. Soavi, M. Mastragostino, Effect of lithium ions on oxygen reduction in ionic liquid-based electrolytes, Electrochem. Commun. 13 (2011) 1090–1093
2- S. Monaco, A. M. Arangio, F. Soavi, M. Mastragostino, E. Paillard, S. Passerini, An electrochemical study of oxygen reduction in pyrrolidinium-based ionic liquids for lithium/oxygen batteries, Electrochim. Acta 83 (2012) 94 – 104
3- A. W. Lodge, M. J. Lacey, M. Fitt, N. Garcia-Araez, J. R. Owen, Critical appraisal on the role of catalysts for the oxygen reduction reaction in lithium-oxygen batteries, Electrochim. Acta 140 (2014) 168-173
4- J. T. Frith, A. E. Russell, N. Garcia-Araez, J. R.Owen An in-situ Raman study of the oxygen reduction reaction in ionic liquids, Electrochem. Commun. 46 (2014) 33–35

More than 20 different ionic liquids were prepared by WWU, with the goal of improving the conductivity and the O2 solubility (and a new, water-based ‘green’ synthetic route has been proposed as well). They were provided to partners upon request for their studies (mainly within WP5, WP6 and WP7). They were thoroughly characterized and these results were published in the following articles by WWU:

5- M. Kunze, E. Paillard, S. Jeong, G.B. Appetecchi, M. Schönhoff, M. Winter and S. Passerini, Inhibition of Self-Aggregation in Ionic Liquid Electrolytes for High-Energy Electrochemical Devices, J. Phys. Chem. C 115 (2011) 19431
6- M. Montanino, F. Alessandrini, S. Passerini, G. B. Appetecchi, Water-based synthesis of hydrophobic ionic liquids for high-energy electrochemical devices, Electrochim. Acta 96 (2013) 124

7- M. Kunze, S. Jeong, G. B. Appetecchi, M. Schönhoff, M. Winter, S. Passerini, Mixtures of ionic liquids for low temperature electrolytes, Electrochim. Acta 82 (2012) 69
8- J. Reiter, E. Paillard, L. Grande, M. Winter, S. Passerini, Physicochemical properties of N-methoxyethyl-N-methylpyrrolidinum ionic liquids with perfluorinated anions Electrochim. Acta 91 (2013) 101– 107
9- J. Reiter, S. Jeremias, E. Paillard, M. Winter, S. Passerini, Fluorosulfonyl-trifluorosulfonylimide ionic liquids with enhanced asymmetry, Phys. Chem. Chem. Phys. 15 (2013) 2565-2571

From these more fundamental studies, parameters were collected by AVL for the modeling and design activities (WP4).

It was also found, as a first step to an operation versus the highly oxidizing O2 cathode, that they could be used versus high voltage oxide insertion cathodes (i.e. charged up to 4.8V) with much improved results as compared with classical carbonate mixtures, which was reported as well by WWU:

10- J. Li, S. Jeong, R. Kloepsch, M. Winter, S. Passerini, Improved electrochemical performance of LiMO2 (M=Mn, Ni, Co)–Li2MnO3 cathode materials in ionic liquid-based electrolyte, J. Power Sources 239 (2013) 490

The performance versus Li metal of IL-based electrolytes, as well as the stabilizing of the Li metal anode (WP5), depending on the electrolyte and separator were studied by WWU. It was found by UNIBO that, unfortunately, the FSI- anion, which allows for highly performing ionic liquids, both in term of conductivity and Li metal cycling was not chemically stable enough for a use in Li-air, which narrowed the choice of anions to perfluorosulfonimide, among which TFSI- allows the best performances in terms of conductivity (although it was shown in [2] that anions comprising more fluorine atoms do increase O2 solubility). New methods for efficiency determination of Li metal electrodes were developed and long term cycling for more than 11 months were reached in liquid IL-based electrolytes. New insight into Li deposition, which occurs behind the protecting SEI, contrary to a popular belief that ‘freshly deposited Li metal would strongly react with O2’, was demonstrated thanks to ex-situ SEM and in-situ optical microscopy studies. It was even possible to acquire videos showing that, if applying a potential of -1V vs Li+/Li do trigger globular dendrite growth, as an obvious result of Li+ full depletion at the interface and the resulting strong electrical field gradients, it is not the case when cycling in conditions at which this full depletion is not expected and that homogeneous plating of 9 mAh cm-2 was possible maintaining a shiny and smooth Li surface. Some of these aspects were (or are about to be) published in:

11- L. Grande, E. Paillard, G.-T. Kim, S. Monaco, S. Passerini, Int. J. Mol. Sci. 2014, 15(5), 8122-8137
12- M. Kirchhöffer, E. Paillard, J. von Zamory, S. Passerini, Influence of separator on hydrophobic ionic liquid-based electrolytes performance
13- L. Grande, J. von Zamory, J. Kalhoff, E. Paillard, S. Passerini, Dendrite-free lithium metal anode: a morphological and electrochemical study using ionic liquid electrolytes (in preparation)

Solid polymer electrolytes were found to be extremely resilient toward Li dendrite growth, even in conditions of current at which almost full Li depletion was expected (i.e. at the limiting steady-state current) allowing the plating of 50 µm of lithium (i.e. a full anode) without dendrite and continuous cycling of Li metal anodes for more than 2800 cycles was achieved as well. Their physico-chemical characterization and use versus Li metal anode (and, in some cases insertion cathodes given that, in a final flow cell, they would, in fine, be used as separator in combination with a circulating liquid electrolyte) were published by WWU in the following:

14- M. Joost, M. Kunze, S. Jeong, M. Schönhoff, M. Winter, S. Passerini, Ionic mobility in ternary polymer electrolytes for lithium-ion batteries, Electrochim. Acta 86 (2012) 330– 338
15- M. Wetjen, G.-T. Kim, M. Joost, M. Winter, S. Passerini, Temperature dependence of electrochemical properties of cross-linked poly(ethyleneoxide)–lithium bis(trifluoromethanesulfonyl)imide–N-butyl-N-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide solid electrolytes for lithium batteries Electrochim. Acta 87 (2013) 779
16- M. Joost, G.-T. Kim, M.Winter S. Passerini, Phase stability of Li-ion conductive, ternary solid polymer electrolytes, Electrochim. Acta 113 (2013) 181– 185
17- M. Wetjen, G.-T. Kim, Mario Joost, G.B. Appetecchi, M. Winter, S. Passerini, Thermal and electrochemical properties of PEO-LiTFSI-Pyr14TFSI-based composite cathodes, incorporating 4 V-class cathode active materials, J. Power Sources 246 (2014) 846
18- I. Osada, J. von Zamory, E. Paillard, S. Passerini, Improved Li-metal/V2O5 polymer battery incorporating crosslinked ternary polymer electrolyte with PYR14TFSI, 271, (2014) 334–341

The investigation of new concepts of artificial protecting SEI were investigated by TAU, in view of stabilizing the Si-based anode prepared by both TAU and KNUTD. A method for the pre-treatment of multiwalled carbon nanotubes (MWCNT) was developed as well as the electrophoretic deposition of composite ceramic-polymer films to potentially serve as SEI. Concerning the Si asnodes, a synthesis of new core-shell silicon-hard carbon nanoparticles attached to MWCNTs was developed by TAU, while KNUTD worked on lower Si content (obtaining excellent cyclability of 700 mAhg-1 anodes)
Finally Li/Si-C-MWCNT cells with PYR14TFSI/LiTFSI electrolyte provided by WWU exhibited reversible capacity 1700 mAh/g (Si) for more than 60 cycles. Some of the results were published in:

19 - R. Blanga, D. Golodnitsky, G. Ardel, K. Freedman, A. Gladkich, Yu. Rosenberg, M. Nathan, E. Peled, Quasi-solid polymer-in-ceramic membrane for Li-ion batteries, Electrochim. Acta 114 (2013) 325– 333
20- V. Khomenko, V. Barsukov, I. Senyk, Electrochemical Properties of Advanced Anodes for Lithium-Ion Batteries Based on Carboxymethylcellulose as Binder, Key Engin. Mater. 559 (2013) 49-55

In parallel, different types of template porous carbons were synthesized within WP6, by UNIBO and CSIC, some of them functionalized with catalysts (many catalysts prepared by KNUTD showed excellent catalytic activity in aqueous media). The electrodes obtained were tested, using IL-based electrolytes and high capacities up to ca. 3000 mAh g-1 with voltage hysteresis ≤ 1V, and reasonable cyclability and efficiency (as compared with state-of-the-art) were obtained without the use of any toxic catalysts, which represent a clear advantage in term of environmental sustainability. Modeling, by CSIC explained different discharge behavior of carbons with different porosity in cells with stirred and unstirred electrolytes. In addition, unlike in aqueous solution, there is no need of inorganic catalysts for O2 reduction in ionic liquid electrolytes, which is advantageous from an eco-design point of view. The following articles report on these advances:

21- V. Khomenko, K. Lykhnytskyi, V. Barsukov, V. Chaban, Composite Catalysts towards Oxygen Reduction in Aqueous Solutions, Key Engineer. Mat. 559 (2013) 57

22- M. Olivares-Marín, P. Palomino, J. M. Amarilla, E. Enciso and D. Tonti, Effects of architecture on the electrochemistry of binder-free inverse opal carbons as Li–air cathodes in an ionic liquid-based electrolyte, J. Mater. Chem. A 1 (2013) 14270-14279
23- M. Olivares-Marín, P. Palomino, E. Enciso, Dino Tonti, Simple Method to Relate Experimental Pore Size Distribution and Discharge Capacity in Cathodes for Li/O2 Batteries, J. Phys.Chem. C 118 (2014) 20772–2078
24- F. Soavi, S. Monaco, M. Mastragostino, Catalyst-free porous carbon cathode and ionic liquid for high efficiency, rechargeable Li/O2 battery, J. Power Sources 224 (2013) 115 - 119

From the automotive requirements, a pack design was developed by AVL (WP4) (who filled 3 patent applications), with an emphasis on energy density, which is the main advantage of the technology. For an automotive use, a power battery has been considered for delivering the peak power (acceleration). As an additional advantage of the circulating electrolyte, it was found that an external O2 harvesting device would act as a heat exchanger for battery cooling. (1-D and 3-D) modeling was done at the cell level, a pack design was developed, and the environmental impact of the Li-air technology was assessed through a Life Cycle Assessment approach by VW.

If harvesting O2 from air (WP7) is still an unresolved issue, the project highlighted the high O2 flow needed for operating a Li-air pack, which confirmed the validity of LABOHR approach of using flowfield plates and the use of high mass loading cathode. A membrane, fully blocking for water, was finally developed by SAES, but the surface area that would have been needed was still unrealistic to integrate it in the final prototype, which finally used a bubbler for saturating the electrolyte with O2.

A first flow cell was demonstrated by UNIBO, showing that forced convection can effectively overcome the diffusion limitation that had been found within WP6:

25- S. Monaco, F. Soavi and M. Mastragostino, Role of Oxygen Mass Transport in Rechargeable Li/O2 Batteries Operating with Ionic Liquids, J. Phys. Chem. Lett. 4 (2013) 1379 -1382

An ancillary prototype for thermal- and electrolyte management was tested and realized by AVL so that flow rate and temperature distribution of the system can be monitored, recorded and evaluated. Finally a 5 cm2 cell, operating with electrodes above 3 mg cm-2 was operated using a circulating electrolyte and an interdigitated flowfield plate was introduced as it was found to be necessary to effectively overcoming most of the O2 diffusion limitation.

One of the highlights of the project was the introduction of a new shuttle concept, by SOTON, published in:

26 – M. J. Lacey, J. T. Frith, J. R. Owen, A redox shuttle to facilitate oxygen reduction in the lithium air battery, Electrochem. Commun. 26, (2013) 74–76

It allows, at the same time short-circuiting O2 solubility and diffusion, and accelerating further the oxygen reduction reaction while solving the electrode pore clogging issue by a delocalized deposition of Li2O2 (WP6) and accelerating O2 transfer to the electrolyte (and thus also addressing O2 harvesting - WP7).

It is worth noting that other laboratories have followed this concept that had a major impact on Li-air research, but that the effective use of shuttles and soluble mediators would, in principle, require the use of a circulating electrolyte, so that to choose the location where the Li2O2 deposition occur as well as to avoid self-discharge of the battery by the soluble species. New redox mediators for improving the charge reaction, as proposed by other laboratories were also proposed by SOTON as well a new shuttles/redox mediator for facilitating the discharge, that are to be published.

Potential Impact:
If the practical use of Li-air secondary batteries is not expected before at least one or two decades, the contribution of the LABOHR project to the field has been major. In particular, the more the project has been progressing the more it appeared that the approach of using ionic liquid-based electrolyte was among the few viable solutions to solve the solvent reactivity issues found with most other solvents, even without considering the volatility issue that only IL address, and many groups have, since then, followed LABOHR approach of using ionic liquids, although most groups are still using ionic liquids of improper purity.

While impressive results have been published outside of LABOHR in high impact factor journals, recent articles [S. Meini, S. Solchenbach, M. Piana, and H. A. Gasteiger, J. Electrochem. Soc., 161(9) (2014) A1306-A1314] suggest that contamination by either water or solvent decomposition product plays a key role in obtaining these results, and that, in fact, these contaminants induce the generation of soluble species, that allow operating air cathode while avoiding (or at least limiting) the electrode clogging issue, in a somehow uncontrolled way. A more elegant approach has been introduced within LABOHR, by the use of soluble mediator or shuttles, for delocalizing the discharge product deposition, which has also been reprised by many laboratories since then. Other promising approach, such as the use of redox mediator, which was proposed by other groups, was also further developed within LABOHR, even though the results are still to be published.

However, similarly to other battery technologies such as Li-S, the presence of soluble species must be addressed and, unless the Li-air cell is engineered (thanks to the control of electrolyte flow, for instance) so that to avoid the shuttling of soluble species between the electrodes and the resulting self-discharge. However, LABOHR paved the way of this field of research that is currently replacing the solid catalyst approach, which, in the case of stable ionic liquid electrolytes, was shown to be ineffective.

As the technological aspects adressed within LABOHR, such as the need for massive O2 flow and of using thick electrodes, as well as the unlikelihood of operating Li-air electrodes in a 3-phase configuration (which would correspond to the results usually reported obtained using ultra-thin electrodes at the interface with an O2 static atmosphere) have been (or are being) disseminated [L. Grande, E. Paillard, S. Passerini, J.Hassoun J.-B Park, Y.-J. Lee, Y.-K. Sun, B. Scrosati, Adv. Mat. (under revision); L. Grande, E. Paillard, Lithium-air Batteries with split O2 harvesting and Redox processes: The main results of the LABOHR project, ISE meeting Lausanne, 04/09/2014; D. Bresser, E. Paillard, S. Passerini, Ionic Liquid-based Electrolytes for Li Metal/Air Batteries: A Review of Materials and the New 'LABOHR' Flow Cell Concept, J. Electrochem. Sci. Tech. 5(2) (2014) 37-44] we expect seeing more laboratories tackling these issues. However, to the best of our knowledge, only LABOHR has been operating Li-air cell in a configuration that could more realistically be extrapolated to a full Li-air pack, even if the current densities and electrodes thicknesses could be increased further.

If the Li-air battery still requires further development and is not expected to reach maturity in a short term future, many results developed within LABOHR can benefit other fields that will most likely find shorter term applications. For instance, the electrolytes and studies concerning Si-based anodes, done within WP5 can benefit the Li-ion batteries field, which is still struggling to use Si-C composites with decent cycling performance.

In addition, the use of Li-metal anode has become more and more popular and many claims have been made concerning the extrapolation of thin solid-state batteries to automotive battery packs, even though it is known neither glass nor ceramic electrolytes possess the elastomeric properties of polymer electrolyte to accommodate the volume changes, which result in contact loss and very limited cyclability, all the more than the volume of cycled Li metal is important. From this point of view, the use of liquid IL-based electrolytes has shown that it is possible to reach long term cycling, not only with FSI-based ionic liquids, but also with TFSI-based ones, providing that the temperature is increased to 60 °C and good performing Li-metal batteries have been prepared within LABOHR using solid polymer electrolytes at temperatures as low as 40 °C (20 °C still allowing the cycling of batteries at very low rate).

The strong decrease of SEI resistance when using some specific ionic liquids and separators could also pave the way to improving further the low temperature performance of Li-metal battery, especially regarding a possible ‘cold start’ by either using liquid electrolytes, or by Li metal pre-treatment and use of solid polymer electrolytes including ionic liquids.

All in all, the impact of LABOHR will probably be first seen in Li-ion and Li-metal battery field, and benefit the development of a greener transportation system that will benefit first from improved Li-ion batteries including Si-C nanocomposites anodes, then we can foresee the evolution of the current Li-metal polymer technology to batteries able to operate at lower temperature, thanks to the advances made in SEI pre-treatments as well as the use of polymer including ionic liquid. More prospective, the use of Li-air cells will most likely require further developments, which include the results obtained within LABOHR, as well as the further development of concepts introduced within LABOHR.

List of Websites:
http://www.labohr.eu

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