NMP3-CT-2006-033181

Ionic **L**iquid-based **Li**thium **Batt**eries

"ILLIBATT"

Specific Targeted Research Project

Integrating and strengthening the European Research Area

PUBLISHABLE FINAL ACTIVITY REPORT

Start of the project: 01/01/2007
Duration: 42 months

Co-ordinator Organisation name: (WWM)
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The ILLIBATT project has four key objectives: (i) development of a green and safe solid-state electrolyte chemistry based on ionic liquids and unique ionic liquid based composites with high performance, (ii) use of novel nano-structured high capacity anodes, prepared with the help of novel ionic liquids, (iii) investigation of the peculiar electrolyte properties and the specific interactions of these electrolytes with advanced commercial and self-prepared electrode (anode and cathode) materials with the goal to understand and improve the electrode and electrolyte properties and thus their interactions, and (iv) construction of rechargeable lithium cells with optimized electrode and electrolyte components. The objectives aim at applications in various cell chemistries, including lithium metal and lithium-ion cells. Furthermore, the developed materials are expected to be useful in an extended range of different cell sizes: from small scale, e.g. micro-batteries, to very large scale, e.g., delocalized storage units (10-20kWh), which could be coupled with small energy production plants based on fuel cells or alternative renewable energy sources, and of course also for various types of electric vehicles (up to 50kWh). The research work in ILLIBATT aims to overcome the well-known technical problems of the present rechargeable lithium battery technology with the goal to:

- perform breakthrough work to position Europe as a leader in the developing field of high energy and environmentally benign and safe batteries and to maintain the leadership in the field of Ionic Liquids;
- develop appropriate solid electrolytes and nano-structured electrode materials which combination allows to realize true solid state lithium batteries;
- develop ***all-solid-state*** concept-cells operating at room temperature with:
  - specific energy higher than 180 Wh/kg with respect to the overall weight of the cell;
  - coulombic efficiency in average higher than 99% during cycling;
  - cycle life of 1,000 cycles with 20% maximum loss of capacity, cycling between 100% and 0% SOC; and
- evaluate their integration in renewable energy sources.

List of participants:

Participation no.	Participant organization name	Org. Type	Participant short name	Country
1	Westfälische Wilhelms University of Münster **Enter date: 01-07-2008**	HE	WWM	Germany
2	Ente per le Nuove Tecnologie, l'Energia e l'Ambiente	RES	ENEA	Italy
3	Center for Electrochemical Technologies	RES	CIDETEC	Spain
4	Chalmers University of Technology	HE	CHALMERS	Sweden
5	Kiev National University of Technologies and Design	HE	KNUTD	Ukraine
6	CEGASA	IND	CEGASA	Spain
7	Sued-Chemie	IND	SC	Germany
8	Photowatt	IND	PW	France
9	Technical University of Graz Institute for Chemistry and Technology of Inorganic Materials (ICTAS) and Institute for Chemistry and Technology of Organic Materials (ICTOS) **Exit date: 30-06-2008**	HE	TUG	Austria

To achieve the key objectives, ILLIBATT project has been organized in 2 Phases:

I. **Materials R&D *(PHASE-1)***
II. **Concept cell realization and testing *(PHASE-2)***

Phase-1 started immediately with the project kick-off, whereas Phase-2 will be put in action after 10 months. As it is necessary to continuously adjust the materials R&D to the progress in concept cell realization and vice versa phase-1 and phase-2 activities have to run in parallel for significant time of the project, in particular in the middle of project duration. In the final 6 months of the project, however, Phase-1 activities will be reduced in order to concentrate the efforts in Phase-2. Figure 1 summarizes the ILLIBATT Breakdown Structure with the indication of the Work Packages (WPs). The interactions between Phase1 and Phase2 are controlled through the WP1 with a continuous day-to-day work of the coordinator, the TACC and the steering committee. The main goal of ILLIBATT coordination activities is to create an equal responsibility and influence on the project outcome for all participants in order to ensure a smooth and regular overall coordination of the project, to reach the fixed scientific and technical objectives in a timely and budget consistent manner with properly reported and disseminated results.

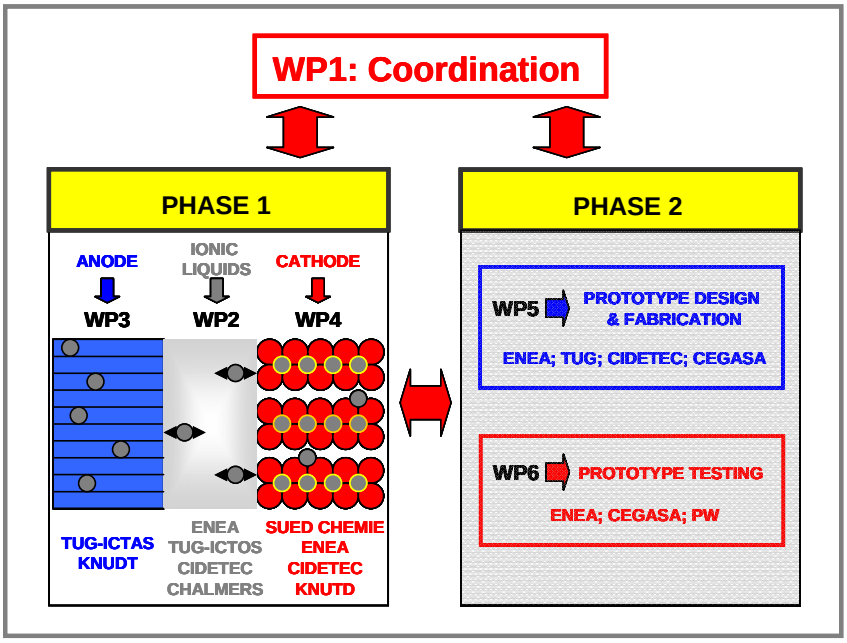

Figure 1: ILLIBATT Breakdown Structure

The major milestone of Phase-1 was the definition of the most promising materials according to the outcomes of WP2-WP4.

The participants cooperatively worked on the synthesis, characterization, testing and optimization of the electrode and electrolyte materials in order to improve the overall technical performance of each single component. The objectives and the main results achieved in Phase-1 are summarized in the following:

Electrolytes R&D

Objectives: Synthesis of ultrapure, anhydrous ionic liquids having a room temperature conductivity of at least 1 mS/cm and preparation of solid state electrolyte materials at the laboratory scale.

Results achieved after 3 years of work:

I. Synthesis of anhydrous (< 1 ppm), high purity ionic liquids (> 99.5 mol.%) with an overall yield close to 90 mol%. The $PYR_{1A}FSI$ ionic liquid exhibits conductivity close to 1 mS cm^{-1} already at -20°C with an electrochemical stability window exceeding 5 Volt.
II. Synthesis of polymeric ionic liquids (PILs) compatible with the ionic liquid (IL).
III. Electrochemical characterization of PIL-based polymer electrolyte. The polymeric electrolyte exhibit conductivity of 10^{-4} mS cm^{-1} with an electrochemical stability window close to 5 Volt, at 40°.
IV. Scaling-up of polymeric electrolyte containing PEO cross-linked, $PYR_{14}TFSI$ and LiTFSI. The polymeric electrolyte exhibit conductivity higher than 10^{-4} mS cm^{-1} at 40°C with very promising mechanical properties.

Anodes R&D

Objectives: Synthesize, select and characterize advanced anode materials (carbons, metals alloys) in the presence of liquid and solid IL-based electrolytes (including nano-structured lithium storage alloys prepared by electroplating).

Results achieved after 3 years of work:

I. Electrodeposition of metals on Copper current collector from IL-based electrolytes at RT.
II. Evaluation of the specific capacity (1145 $mAhg^{-1}$) of Silicon electrodes in combination with ILs at RT.
III. Evaluation of commercial graphite/carbonaceous materials in IL based electrolyte systems.
IV. Evaluation of the performance of $Li_4Ti_5O_{12}$ electrodes containing fluorin-free binder (CMC) in IL based electrolyte.

Cathodes R&D

Objectives: Selection and Optimization of an appropriate cathode material for the Ionic-Liquid based Solid-State Electrolyte Battery

Results achieved after 3 years of work:

I. Evaluation of the performance of $LiFePO_4$ electrodes containing fluorin-free binder (CMC) in IL based electrolyte.
II. Fabrication and testing of composite $LiFePO_4$ cathodes tapes, using PEO-LiTFSI-ILs membranes, as electrolyte separators.
III. Fabrication and testing of composite $LiFePO_4$ cathodes tapes, using PIL-LiTFSI-ILs membranes, as electrolyte separators.

The main objectives in the Phase-2 are the design and fabrication of solvent-free, all solid state concept cell batteries and the evaluation of the electrochemical and safety performance of the all solid state concept cell battery. As the proposed materials are to a substantial part highly innovative and thus not thoroughly characterized and tested, activities of phase-2 started on month 10 of the project.

Concept Cell Design and Fabrication

Objectives: Design and fabrication of solvent-free, all solid state concept cell batteries.

Results achieved after 3 years of work:

I. Design of all solid state concept cell lithium batteries: stacked bipolar cells in aluminium soft package.
II. Fabrication of prototypes $Li_4Ti_5O_{12}$ / 0.1LiTFSI – 0.9 $PYR_{14}FSI$ / $LiFePO_4$
III. Fabrication of prototypes Li/ PEO-cl/ $LiFePO_4$
IV. Fabrication of prototypes Li/ PIL / $LiFePO_4$

Concept Cell Testing

Objectives: Evaluate the electrochemical and safety performance of the all solid state concept cell battery.

Results achieved after 3 years of work:

I. Definition of the electrochemical testing procedure to be performed on the concept cells, taking into account PV application and its focus on reliability rather than power.
II. Definition of reliability and safety tests suitable for the concept cell batteries.
III. Testing of ILLIBATT prototype: cycling stability, temperature behaviour, Safety test.