

Development of a new Li metal-Gel Polymer Electrolyte thin flexible battery design operating at room temperature

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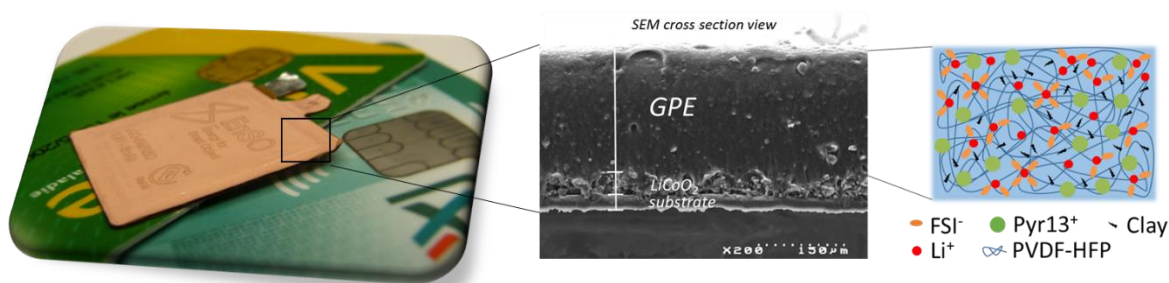
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Graphical Abstract



Abstract

In this study, we develop a new Li-metal battery design merging with the IoT requirements, mainly the low thickness, the thermal stability and the flexibility. To reach these specifications, we firstly synthesized an efficient gel polymer electrolyte (GPE) composed of a PVDF-HFP polymer network, a LiFSI: Pyr₁₃FSI ionic liquid binary solution and a lithium montmorillonite

LiMMT clay. The as-synthesized material exhibits a high ionic conductivity (0.48 mS cm^{-1} at 25°C) and a good thermal stability, up to 140°C . In parallel, a new battery design with an optimized ratio of packaging to active material thickness is developed. In this design, copper foils act both as current collector and as battery casing, decreasing the overall cell thickness. Li metal batteries are realized using this new design and the developed GPE material. The cell thickness is 360 and $760 \mu\text{m}$ for single side and double-sided architectures respectively. These batteries show well functioning under high bending and exhibit a good cycling ability with a remaining capacity higher than 85% after more than 200 cycles at 25°C . Thanks to the combination of the original Cu packaging and the flexible GPE membrane, developed Li-metal batteries exhibit promising properties to merge with the new IoT requirements.

1. Introduction

The 2019 Chemistry Nobel Prize was attributed to three researchers, J. Goodenough, S. Whittingham and A. Yoshino, who contributed to the enhancement and the advent of the lithium ion batteries [1, 2]. These developments modify our society since the 1990s and the first commercialization of a lithium ion battery by Sony Corporation [2]. The development of lithium ion batteries then became exponential with its integration in numerous products, from the electric vehicles to the inevitable smartphones. More recently, the emerging of Internet of Things (IoT) provides a great easiness in everyday life fields such as smart homes, wearables, medical devices, data security, etc. The number of connected IoT devices was multiplied by 3 since the 2010s [3]. These new applications lead to toughen battery requirements like flexibility, limited thickness ($< 500 \mu\text{m}$), etc. The pouch cell configuration is the only current

commercial one that could fit with these new requirements. However, despite a smaller dead volume than the cylindrical or prismatic geometries [4], if very low thickness are targeted (< 500 μm), the classical pouch cells still suffer from an important part of the total thickness dedicated to the packaging. Furthermore, IoT applications require an enhanced security (medical devices, battery-user proximity etc.) [5] and currently, the main safety hazard originates from the electrolyte leakage and its irreversible decomposition. Hence, lithium polymer batteries are studied since the 1970s with the solid polymer electrolyte (SPE) described by Armand *et al* [6-8] composed of a solid polymer matrix of crystalline poly(ethylene) oxide (PEO) and a lithium salt. It could also enable the use of Li metal anode, considered as the ideal anode material regarding its high theoretical capacity (3860 mAh g^{-1}) and low redox potential (-3.04 V vs NHE) [9]. Bolloré, with the BlueCar and Autolib car sharing development in Paris, brought lithium metal-polymer (LMP) batteries to a production level [10]. Nevertheless, the main drawback of these electrolytes remains its low ionic conductivity at room temperature (10^{-4} - 10^{-7} S cm^{-1}) [11, 12]. These restrictions limit the C-rate and/or impose a working temperature around 60-80°C. Indeed, the PEO matrix, despite a high dielectric constant ($\epsilon = 8$) which enhances the salt dissolution, suffers from a limitation of Li mobility. The Li is crown solvated by oxygens of the polymer chains whose motion is restricted by the polymer crystalline domains [13]. Several studies are working on improving performances by limiting the host polymer crystallinity for example with the addition of a plasticizer [14, 15], ceramic fillers [16, 17] or nano-clay [18, 19]. Among these nano-clays, the lithium montmorillonite (LiMMT) presents various interests like its low cost, its ability to transfer Li^+ and its layered structure which can intercalate polymer chains [18, 20]. The added plasticizer can also be a liquid electrolyte which, trapped into the host polymer network, increases the ionic conductivity as initially proposed by Feuillade *et al* [12, 21, 22]. The Li salt

dissolution is then not necessarily provided by the polymer host and new polymer matrix can be investigated. For example, the poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP), widely used as binder in electrode composition, is not involved in the Li solvation and is studied as GPE host due to its amorphous domains able to trap large amounts of liquid phase without mobility restriction [14, 22, 23]. To enhance safety and thermal stability, many groups started to use ionic liquids as solvent instead of standard carbonate ones due to their low vapor pressure and high thermal stability [24-26]. Ionic conductivities around 1 mS cm^{-1} can be achieved with these ternary electrolytes. The added lithium salt also plays an important role on the electrochemical performances. Nowadays, the salt used in commercial Li batteries is predominantly the LiPF_6 but its low thermal stability ($\sim 55^\circ\text{C}$) and poor resistance against chemicals and moisture motivated the development of new lithium salts. The most studied are the lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) and the adjoining lithium bis(fluorosulfonyl)imide (LiFSI) which shows a higher conductivity in solution and a better compatibility with Al current collector in the high potential region [27-29]. For many years, low salt concentrations, inducing higher conductivities due to lower viscosities, have been considered as favorable [30, 31]. However, an increasing number of recent studies reports the positive effect of higher salt concentration, mainly on the Li transference number and molecular interactions [32-35].

In this work, we report on the synthesis of a new gel polymer electrolyte performing at room temperature. The GPE material is characterized from a physico-chemical and electrochemical point of view. Simultaneously, we develop an innovative battery design with an improved ratio of packaging to active material thickness. The synthesized GPE is finally integrated in a full LiCoO_2/Li battery using the new battery design.

2. Experimental

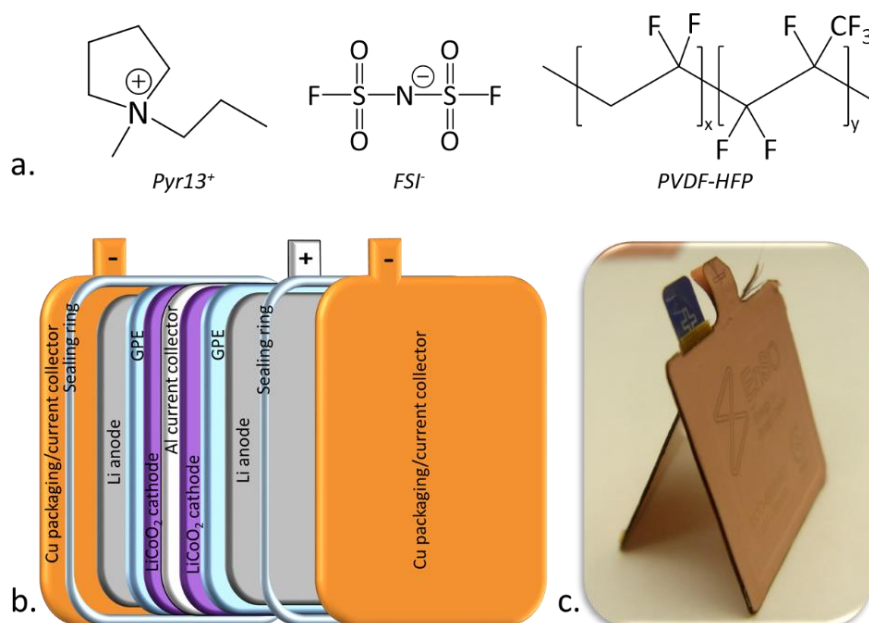


Figure 1: a) Chemical Structures of Pyr₁₃⁺ cation, FSI⁻ anion and PVDF-HFP host polymer; b) Schematic exploded view of the new battery design (double-sided configuration); c) Image of the as-realized batteries.

2.1. Materials and samples preparation

Lithium bis(fluorosulfonyl)imide (LiFSI, > 99.9 %) is purchased from Suzhou Fluolyte Co. N-propyl-N-methylpyrrolidinium bis(fluorosulfonyl)imide (PYR₁₃FSI, 99.9 %) are purchased from Solvionic (Figure 1a). Cloisite Na⁺ (NaMMT) is purchased from BYK-Chemie. Poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP, SOLEF 21216) is kindly supplied by Solvay.

The GPE electrolyte is composed of a binary liquid solution (PYR₁₃FSI and LiFSI), a PVDF-HFP as host polymer and a phyllosilicate clay as reinforcing filler. Firstly, the liquid binary solution, consisting of LiFSI dissolved in PYR₁₃FSI, with a LiFSI: PYR₁₃FSI molar ratio of 1.1: 1, is homogenized for 1 h by magnetic stirring. PVDF-HFP, clay and acetone (VWR, NORMAPUR, > 99.8 %) are then added to the solution which is stirred during 1 night. Typical proportions are 1.2 g PVDF-HFP, 1.68 g clay, 4.8 mL liquid binary solution (LiFSI + PYR₁₃FSI) and 90 mL acetone.

The pristine clay (Na-MMT) is previously modified by a lithiation process [18, 36]. The ion-exchange Li-MMT clays are prepared by cationic exchange between the sodium cations of the montmorillonite clay platelets and lithium cations. 30 g of Na-MMT are dispersed in 3 L of distilled water added with 60 g of LiNO_3 (Honeywell, >98%). The solution is stirred for two days at 50 °C in order to exchange the cations and then centrifuged to separate the Li-MMT clay from the supernatant containing the excess ions. To purify the clay, Li-MMT is redispersed in distilled water (3 L) and once again centrifuged. The modified clay is then dried under vacuum at 80 °C during one night.

The GPE membrane is then deposited by spray technique. The substrate is aligned on a plate heated at 80 °C. The flow rate through the spray nozzle is fixed at around 10 mL min⁻¹, the speed of the spray head at 40 cm s⁻¹ and the atomization is obtained thanks to two lateral air jets with a pressure of 7 bar. The spray coating is realized by alternating deposition and drying steps (20 s). These steps are repeated until a deposited mass of about 40 mg cm⁻² is achieved which corresponds to about 80 to 100 deposition steps. To limit the clay decantation prior to spray, a fluid dispensing system with a reduced tank volume (10 mL) is used. Thus, this tank is refilled two times during the GPE deposition. At the end of the process, GPE films are dried under vacuum at 45 °C during one night.

For material characterizations (SEM images, Arrhenius plots, etc.), standalone membranes are formed by deposition on aluminum foil following by a GPE detachment. The obtained membranes are between 250 and 300 μm thick.

For battery integration, GPE is directly deposited on cathode material. The cathode consists of a LiCoO_2 coating (96 wt% of LiCoO_2 , 2 wt% of carbon and 2 wt% of PVDF) deposited on

aluminum foil by standard doctor blade method. The electrode loading is about 14 mg cm^{-2} which corresponds to a theoretical capacity is 1 mAh cm^{-2} . The GPE/LiCoO₂/Al foil stack is then cut into 19×19 mm samples to be integrated in the new battery design.

2.2 New battery design description

The new battery design is mainly distinguished by the use of a copper foil acting both as current collector and as packaging allowing the overall battery thickness decrease. This new battery design is described in Figure 1b. The design can be realized in single side or double-sided architecture depending on the intended battery thickness and capacity. The battery is composed of Cu foils (12 μm , 25×25 mm out of connection tabs) and sealing rings (Tesa glue, 115 μm , 25×25 mm external size and 20×20 mm internal aperture) acting as airtight packaging. Sealing rings are fixed on Cu foils by thermal curing (120 °C). To improve the design reliability and avoid short circuits during the sealing process, a polypropylene ring (25 μm , 21.5×21.5 mm external size and 13×13 mm internal aperture, not represented in Figure 1b) is added on the sealing ring. Li anode (Albemarle, 50 μm , 17×17 mm) is then pressed on the Cu foil in the center of the sealing ring. Two previously prepared GPE/LiCoO₂/Al foil samples are aligned back to back and sandwiched between two Li anode/sealing ring/Cu foil stacks. The assembly is then vacuum packed at 120 °C to ensure a good anode/cathode contact and achieve the airtight. The obtained batteries are shown in Figure 1c. The thickness is about 400 μm for the single side architecture and 600-750 μm for the double-sided ones. The overall size is 25×25 mm (out of connection tabs) with 2.89 cm^2 of active material surface.

2.3 Morphological, thermal and mechanical characterizations

The surface morphology and thickness of the GPE are observed using a Hitachi 4100 scanning electron microscope (SEM).

Thermogravimetric analyses (TGA) were carried out under air atmosphere using a Setaram SetSys equipment for GPE material or argon atmosphere using a Netzch STA449 F1 Jupiter equipment for the neat material. Thermal gradient is fixed at 5 °C min⁻¹. The degradation temperature (T_d) is defined as the temperature of 1 wt% loss for the considering degradation. The onset temperature (T_{on}) is defined as the point of intersection of the slopes at the beginning and the end of the degradation as described in Figure S1 (supplementary file).

2.4 Conduction and Electrochemical measurements

Ionic conductivity is determined by electrochemical impedance spectroscopy (Biologic VMP3 multichannel galvanostat/potentiostat, frequency range : 100 mHz-1 MHz, $\Delta V = 10$ mV) on coin cells composed of two stainless steel disks separated by a 200 μ m thick self-standing GPE membrane (16.5 mm diameter). Measurements are done in a temperature controlled climatic chamber between 25 and 80 °C (298-353 K). Due to the presence of the ionic liquid in the GPE, the ionic conductivity is increased and the Nyquist plot did not display a semi-circle. The Nyquist plots are then adjusted with a simple equivalent circuit model composed of a resistance (R) and a constant phase element (CPE) in series representing the electrolyte resistance (R_{el}) (connection resistances being neglected) and the blocking electrodes respectively.

Cycling ability is evaluated on Li symmetrical coin cells. Li electrodes (135 μ m, Albemarle) and GPE membranes are cut into 12 mm and 14 mm disks respectively. Cells are cycled at various current densities from 0.04 to 0.4 mA cm⁻² using a Biologic VMP3 multichannel

galvanostat/potentiostat at room temperature, 100 cycles for each current density. The electrochemical performances of LiCoO₂/GPE/Li stack are firstly evaluated on coin cell configuration. In this case, the diameter of Li anodes and LCO cathodes is 15.5 and 14 mm respectively. The electrodes are sandwiched between two stainless steel disks (15.8 x 0.5 mm) and the overall system is sealed into a coin cell (GELON, SS316) under a pressure of 500 psi.

New battery design cycling ability is determined by long term cycling with potentiostatic charge at 4.2 V during 2 h and galvanostatic discharge at various currents (from 1 to 4 mA) using a Biologic VMP3 multichannel galvanostat/potentiostat. Electrochemical impedance spectrum are recorded at 4.2 V for each cycle (100 mHz-1 MHz, $\Delta V = 10$ mV). Characteristic frequency and resistance of each contribution are graphically determined.

3. Results and Discussion

3.1 GPE characterizations

The thermograms of the as synthesized GPE and neat materials are shown in Figure 2a, related degradation and onset temperatures (T_d and T_{on}) in Figure 2b. Considering that the GPE characterization is done under ambient atmosphere, the first loss below 100 °C is attributed to the water adsorbed during sample preparation and characterization and is not relevant. The first significant loss occurs around 141 °C and is attributed to the beginning of the LiFSI decomposition. In the literature, the thermal behavior of the LiFSI salt is still deeply discussed. Its degradation temperature generally varies from 160 to 200 °C depending on the determination method (1 wt% or 5 wt% mass loss considering, static or dynamic thermal conditions, etc.) [29, 37]. Kubota *et al* [38] even reports a degradation temperature lower than 70 °C under static conditions. Beyond the determination methods, these variations are also

attributed to composition and purity variations. The thermogram of our neat LiFSI (dash orange curve in Figure 2a) exhibits a two stages decomposition with a first loss at 179 °C and a second one around 308 °C (T_{on}), close to those observed by Kerner *et al* [37] and also evidenced in the liquid solution curve (dash dot green curve in Figure 2a). The LiFSI decomposition temperature in the GPE is slightly lower than the neat material one (179 °C) but close to the one observed in the liquid solution Pyr₁₃FSI+LiFSI (140 °C). However, the decomposition kinetic seems to be increased since the mass loss occurred in a restricted temperature range. This could be explained by a confinement effect that leads to a liquid repartition in many little volumes split in the polymer matrix instead of a large unitary volume for the neat materials (LiFSI or liquid solution). The second major mass loss in the GPE thermogram at 266 °C is attributed to the second step of the LiFSI degradation and the Pyr₁₃FSI decomposition. The degradation temperature is also slightly decreased compared with the neat Pyr₁₃FSI one (291 °C). The last mass loss at 431 °C corresponds to the PVDF-HFP decomposition. This value is close to the one reported in the literature [39]. The exact mass loss value of each decomposition step is hardly readable due to the superposition of the degradation phenomenon. However, at 600°C, the remaining mass is about 14 wt%.

SEM cross section view of the GPE deposited on a LiCoO₂ cathode coating is presented in Figure 2c. The GPE is 290 µm thick formed by three distinct layers of about 90-100 µm. Each layer corresponds to the three spray deposition stages required by the tank refill as explained in the experimental part. A higher magnification focused on the GPE material is presented in the Figure 2c inset. It shows the presence of small particles (500-800 nm) which may corresponds to clay aggregates. The material seems to be dense within the layers but exhibits some porosity at layer interfaces.

The ionic conductivity measured at 25°C is 0.48 mS cm⁻¹ which is quite good compared with the literature. The Arrhenius plot (Figure 2d) exhibits a linear shape (R^2 0.997) corresponding to the Arrhenius behavior described by the following equation:

$$\sigma = Ae^{\frac{-E_a}{RT}}$$

With A the pre-exponential factor, R the perfect gaz constant and E_a the activation energy (J mol⁻¹). This behavior is quite different from the expected one as a GPE usually exhibits a Vogel-Tammann-Fulcher behavior based on the free volume theory [40, 41]. It can be explained by the high liquid phase content of our GPE where the Li mobility is not directly correlated to the segmental motion and mainly thermally activated. The addition of LiMMT clay, which provides an additional diffusion path for lithium, can also contribute to this unexpected behavior.

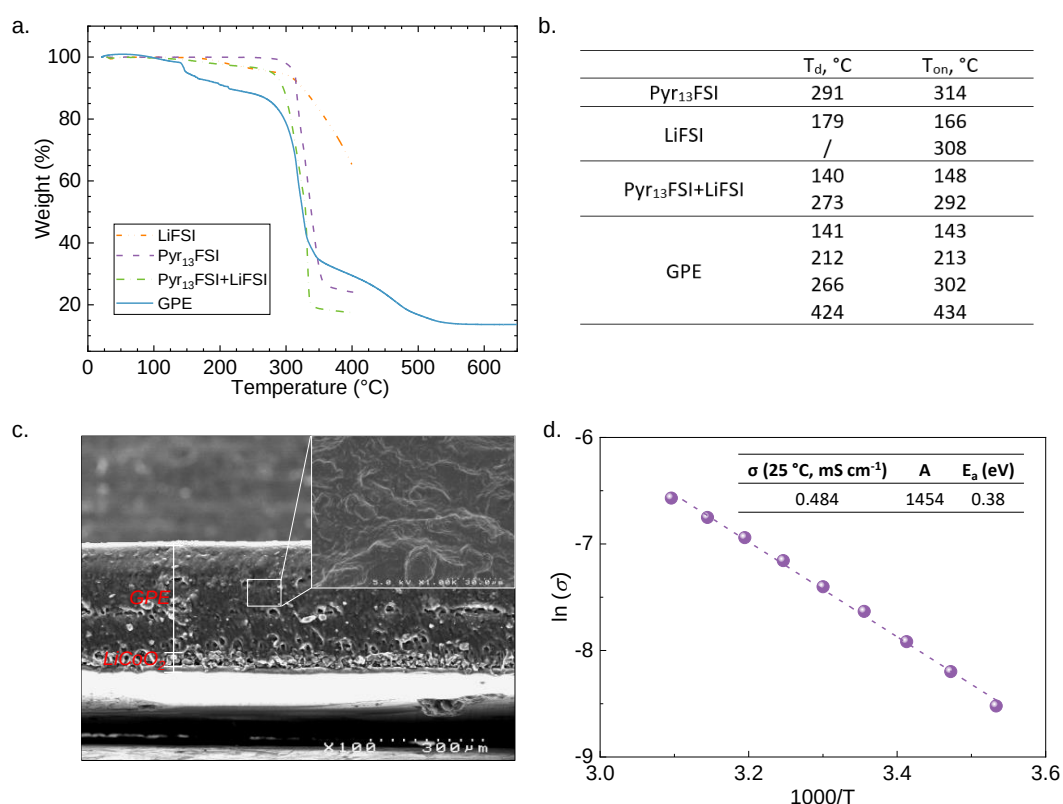


Figure 2: TGA of the neat materials and the as-synthesized GPE (a) and related degradation and onset temperatures (b); cross section SEM view of a GPE/LiCoO₂/Al stack (c); Arrhenius plot and fitting of the as-synthesized GPE (d).

3.2 GPE electrochemical ability

The voltage-time profiles of a Li symmetrical cell integrating the GPE is shown in Figure 3a and the associated reached potential for each cycle in Figure 3b. For the lower current densities (0.04 and 0.08 mA cm⁻²), the maximum potential remains relatively stable upon cycling and a steady state is achieved at the end of each cycle. When the current density increases (0.4 mA cm⁻²), the steady state is no more achieved (no plateau at the end of each cycle) and the voltage-time curve shape is modified throughout cycling. An additional overpotential appears (inset in Figure 3a) indicating a dendritic lithium morphology as previously described by Best *et al* [42]. The dendrites formation increases the electrode surface area. This surface modification decreases the current density resulting in a maximum potential decrease evidenced in Figure 3b. The galvanostatic curve of the GPE integrated in a LiCoO₂/Li cell is presented in Figure 3c. A slight polarization is visible at the beginning of the galvanostatic discharge. This polarization can be attributed to the relaxation after the 4.2 V constant voltage charge release and the GPE thickness (~ 300 μm). It does not allow the detection of the two LiCoO₂ characteristic plateaus at high potential but the discharge curve shape remains close to the expected one for a LiCoO₂ material [43, 44]. The first discharge capacity is 1.06 mAh cm⁻², equal to the expected one. It drastically falls in the first cycles to stabilize around 0.5 mAh cm⁻² and then slowly increases in the next cycles to reach 0.8 mAh cm⁻² after 800 cycles. The discharge current of 0.35 mA is close to the current at which dendritic growth is observed during Li/Li cycling tests (Figure 3a). This dendritic growth phenomena can be amplified by the coin cell geometry. Indeed, the GPE membrane is quite thick (~290 μm) and assumed to be highly pressed during the sealing of the coin cell. Its properties may be altered by this substantial compression. However, despite this unusual evolution, the system is able to

achieve more than 800 cycles with a remaining capacity of more than 80 % of the initial one at room temperature that is quite promising. This high compression issue can be confirmed and solved by the integration in our new battery design.

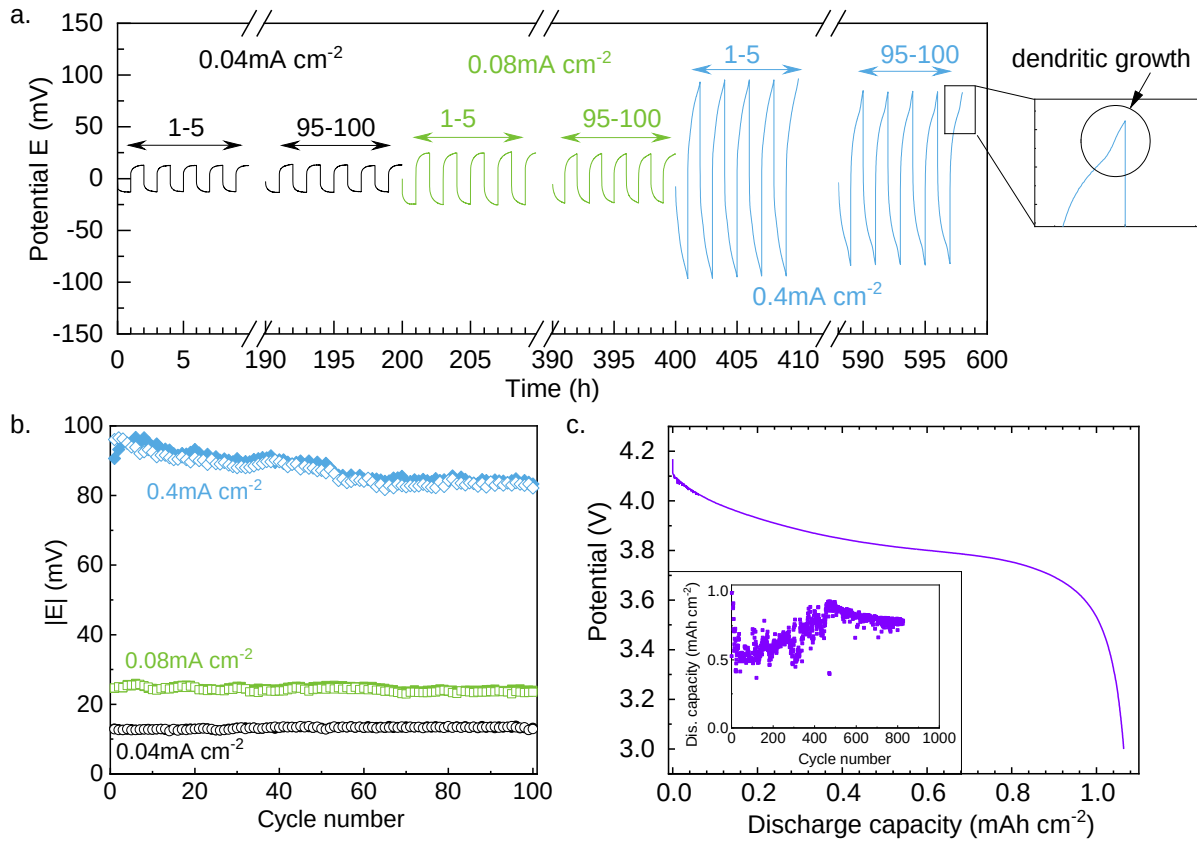


Figure 3: Voltage-time profiles of Li symmetric cell integrating GPE, 100 cycles at 0.04, 0.08 and 0.4 mA cm⁻²(a) and associated maximum reached potential at each cycle for the various current densities (b); Discharge curve of a LiCoO₂/GPE/Li coin cell and cycling ability upon potentiostatic charge 4.2 V 2 h and galvanostatic discharge 0.35 mA cm⁻² (c).

3.2 GPE integration in new battery design

The GPE is integrated in the new Li metal battery with a LiCoO₂ cathode material. The overall battery thickness is low, as shown in Figure 4a compared to a 2 cts coin, 360 μm and 760 μm for the single side and double-sided architectures respectively. The association of the GPE and copper new battery packaging allows a high flexibility and this new battery design still works under high bending constraint as seen in Figure 4b.

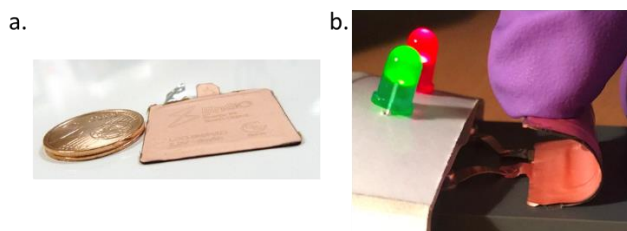


Figure 4: Single-side new battery design beside a 2 cts coin (a); Double-sided battery working under high bending constraint (b).

Double-sided discharge galvanostatic curves for various currents are shown in Figure 5a. The polarization increases with the discharge current involving a lowering of the potential plateau. The potential drop at the beginning of the discharge linearly increases with the current from 150 mV to 250 mV for 1 mA and 4 mA respectively. These observations are in good accordance with the important GPE membrane thickness previously observed ($\sim 290 \mu\text{m}$). However, up to 2 mA, this polarization increase has only a poor impact on the total discharge capacity. Cycling ability at 25°C is presented in Figure 5c. For the single-side architecture, the discharge capacity is quite stable upon cycling and the remaining capacity is 2.73 mAh after 200 cycles, *i.e.* 86 % of the maximum capacity and 103 Wh L^{-1} . The double-sided one is initially cycled with a discharge current of 1 mA. The initial capacity is 5.76 mAh (96 % of the theoretical capacity, 103 Wh L^{-1}). The discharge capacity slowly decreases in the first 40 cycles up to 5 mAh. At 40 cycles, the discharge current is set to 2 mA. This current increase seems to stabilize the discharge capacity, which remains quite constant in the next cycles. The remaining capacity is about 5.11 mAh after 200 cycles, *i.e.* 87 % of the maximum capacity and 91 Wh L^{-1} . Cross section views of the GPE before and after cycling are shown in Figure 5b. After cycling, the GPE morphology is modified with the formation of flakes, not evidenced in the initial sample (before cycling). Moreover, there is less load effects during the SEM observations meaning that electrons removal is easier and indicating a higher electronic conductivity. This result might be a result of the Li dendritic growth and is consistent with the better cycling ability at

higher current considering that the current increase hides the electronic conductivity rise. However, this electronic conductivity rise is detrimental for the long term cycling and the coulombic efficiency. The evolution of the impedance spectra is shown in Figure 5d. The spectra shape of single-side and double-sided architectures are close. The R_0 value is attributed to the electrolyte resistance (contact resistance being neglected) and slowly decreases upon cycling. This decrease can be attributed to a surface increase resulting from a lithium dendritic growth as predicted by the Li symmetrical cell cycling. The first semi-circle in the middle high frequency domain is slightly flatten, mainly for the single-side architecture, and can comprise two contributions. In a first approach, we will consider it as a single one. For the single-side architecture, the characteristic frequency (f_c) is 8.9 kHz, close to the 8 kHz reported in the literature for a polymer-Li interface [45]. For the double-sided architecture, the characteristic frequency (f_c) is largely decreases (1.9 kHz) indicating that the formed interface is quite different from the single-side architecture one, f_c being independent of the surface and the thickness. The same observation is done for the second contribution in the medium frequency area with a decrease of the f_c in the double-sided configuration (from 54 to 11 Hz). This loop represents the charge transfer of the LiCoO_2 charge state. For both architectures, this second contribution tends to disappear with cycling progress but the f_c remains quite constant. The first contribution corresponding to the Li/GPE interface increases upon cycling and a new contribution appears in the high frequency region. This contribution has been previously observed in a configuration without Li metal anode ("Li-free") and is attributed to the LiCoO_2 /GPE interface (Supplementary file Figure S2). It means that the LiCoO_2 /GPE interface is formed and/or modified upon cycling and its resistance increases either by a thickness increase or a composition modification. Post-mortem analyses by XPS are undergoing to confirm this hypothesis.

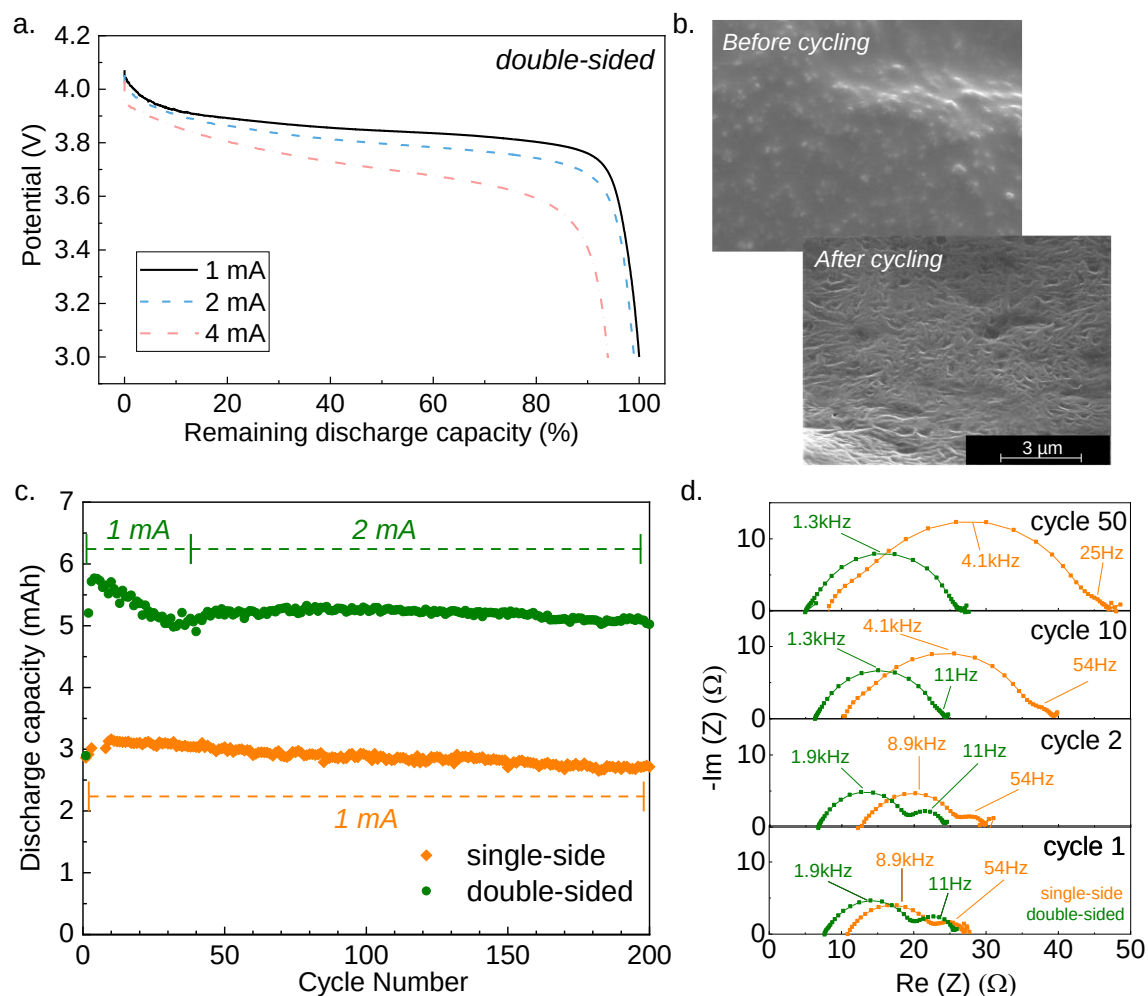


Figure 5: Galvanostatic discharge curves of a double-sided architecture for various currents (25 °C) (a); Cross section SEM images of a GPE before and after cycling (b); Cycling ability of LiCoO₂/GPE/Li new design batteries in single-side (orange curves) or double-sided (green curves) architectures (charge at 4.2 V 2 h, galvanostatic discharge at 1 mA, 25 °C) (c) and charge state (4.2 V) batteries impedance spectra at various cycles (d).

4. Conclusion

The studied GPE, composed of PVDF-HFP, LiFSI, Pyr₁₃FSI and LiMMT clay, exhibits a high thermal stability up to 140 °C and an ionic conductivity of 0.48 mS cm⁻¹ at 25 °C, close to the state of the art for a GPE integrating ionic liquid based solvent. The discharge capacity of a LiCoO₂/GPE/Li stack integrated in a coin cell system is 1 mAh cm⁻², equal to the theoretical one and remains high after more than 800 cycles (> 80 % of the initial capacity). However, the capacity evolution upon cycling is quite unusual with many disparities attributed to the high compression in the coin cell configuration, amplified by the important GPE thickness (290 μm).

The new Li-ion metal battery design avoids this phenomenon. The dual role of the copper foil, acting both as a current collector and a casing, allows to drastically decrease the part of the packaging in the overall cell volume compared to standard pouch cell architecture. Thus, the total thickness of these cells is 360 and 760 μm for single-side and double-sided architectures respectively, GPE representing a large part of this thickness. This design is flexible and exhibits a good functioning under high bending constraint. The cycling ability is good with a capacity retention higher than 85 % after more than 200 cycles regardless of the architecture. SEM and EIS show an evolution of the membrane upon cycling suggesting a detrimental evolution of the GPE electronic properties and GPE/electrode interfaces. Studies are then in progress to improve the system by optimizing the GPE composition to stabilize the interfaces and its electronic properties and reducing its thickness while preserving its mechanical and electronic properties that allow it to act as a separator. Nevertheless, this new battery design integrating an original Cu packaging and flexible GPE exhibits promising properties to merge with the new IoT specifications.

Supporting Information Available

Supplementary information are available to complete results presented in the manuscript: ATG thermogram, electrochemical impedance spectrum.

Acknowledgements

Authors thank Florence Rouillon and Benjamin Amestoy (CEA/LITEN/DEHT) for their technical expertise on electrode fabrication process and access to their facilities.

Financial support for this work was provided through the EnSO project. EnSO has been accepted for funding within the Electronic Components and Systems For European Leadership Joint Undertaking in collaboration with the European Union's H2020 Framework Program (H2020/2014-2020) and National Authorities, under grant agreement n° 692482.

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