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# Computer simulation evaluation of the geometrical parameters affecting the performance of two dimensional interdigitated batteries

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ABSTRACT

The effect of the geometrical parameters of interdigitated batteries, including the number, thickness and the length of the digits, on the delivered battery capacity was investigated in order to optimize battery performance. This optimization was carried out in two dimensions maintaining the area of the different components constant and based on the Doyle/Fuller/Newman theoretical model that describes the electrochemical and ionic transport processes within separator and electrodes. It is observed that the delivered capacity of the battery increases with increasing number of digits as well as by increasing the thickness and the length of the digits, which is related to the smaller path for lithium ion movement during the intercalation/deintercalation processes. Thus, an optimized interdigitated geometry with eight digits shows a delivered capacity of 323 $Ah \cdot m^{-2}$ at 350 °C in comparison with 0.7 $Ah \cdot m^{-2}$ for a conventional geometry.



## 1. Introduction

Lithium-ion batteries are nowadays the most relevant and efficient energy storage systems, increasingly used for applications in portable electronic products, such as mobile-phones, computers, e-labels and disposable medical testers, hybrid electric vehicles (PHEVs) and electric vehicles (EVs) [1].

The rechargeable battery market is expected to reach $22.5 billion dollars and the growth of the lithium- ion battery market in 2016 is expected to reach 25%. The increasing demands of the automotive and mobile phone sectors result in an increasing need for lithium ion battery autonomy, power and capacity [2].

The widespread presence of lithium-ion batteries is due to their advantages in comparison with other battery systems, as they are lighter, cheaper, have higher energy density (between 100 and 265 Wh $kg^{-1}$), lower self-discharge, no memory effect, prolonged service-life and higher number of charge/discharge cycles [3,4]. Improving lithium-ion battery performance is nevertheless needed with respect to specific energy, power, safety and reliability [4].

Typically, the performance of a battery is optimized for either power or energy density by modifying the chemistry and materials for electrodes (anode and cathode) and separators in conventional two-dimensional structures [5–7]. This structure is defined as a layer-by-layer configuration such as cathode/separator/anode [8].

Nevertheless, this structure is limited by the slow transport of lithium ions and hindered accessibility to the material at the back of the electrode, close to the current collector [9].

Taking this limitation into account and in order to maximize power and energy density, interdigitated structures are being developed [9]. The interdigitated geometry consists of electrode arrays of rods separated by a solid electrolyte, i.e., lithium salts put directly into the polymeric matrix without organic solvent present in electrolyte. In this way, the surface area of the electrodes increases without additional side reactions on the electrode surfaces [10].

This configuration leads to shorter $Li^+$ transport paths, reducing ion diffusion lengths and electrical resistance across the entire battery system, as well as to higher energy density of the cell within the same areal footprint [9–11].

In this context, interdigitated batteries using high capacity manganese oxide cathodes and lithium anode have achieved a capacity of up to 29.5 μ Ah/$cm^2$, which is 10× the average capacity of rechargeable conventional batteries [12].

Three dimensional (3D) interdigitated architectures have been fabricated by printing concentrated LFP-LTO based inks, showing a high areal energy density of 9.7 J $cm^{-2}$ at a power density of 2.7 mW $cm^{-2}$ [13].

3D printing was also used for the fabrication of batteries based on $Li_4Ti_5O_{12}$ (average particle diameter of 50 nm) and $LiFePO_4$ (average particle diameter of 180 nm). This battery (960 μm × 800 μm, electrode

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width = 60 μm, spacing = 50 μm) shows a high areal energy density of 9.7 J $cm^{-2}$ at a power density of 2.7 mW $cm^{-2}$ [13].

The interdigitated architecture mostly depends on the aspect ratios (length/width) that can be achieved as well as on the geometry of the electrode. In this way, computer simulations of battery performance are important and critical for evaluating the optimized geometries before experimental implementation [14,15].

In order to simulate battery operation, the couplings of different physical-chemical levels are needed. Macroscopic models allow geometrical and dimensional optimization of the battery components and mesoscale models are suitable for understanding and improving the different components of the battery: physical-chemical properties of the materials to be used as electrodes and separators and the choice of the most suitable organic solvents for electrolytes [15–17]. Theoretical simulations on 3D battery architectures have been addressed by focusing on determining an optimal electrode cylinder array configuration [18] as well as the planar tessellated electrode geometry of square and circular electrode arrays, in which the cell capacity can be increased by simply adding more electrodes in the plane of the array or increasing the height of the electrodes [19]. Further, a Finite Element Analysis (FEA) electrochemical model has been developed for several of the main 3D battery architectures such as interdigitated cylinders, concentric cylinders and interdigitated plates using a non-porous electrode (particle-scale) electrochemistry model [20]. The effect of the solid electrolytes ionic conductivity was also analysed for interdigitated structures, the discharge capacity increasing with increasing of ionic conductivity [21,22].

Theoretical simulation was also used to demonstrate that the electrode thickness can significantly influence many key aspects of a battery such as energy density, temperature response, capacity fading rate and overall heat generation, among others [23].

In 3D pillar structures, template pillar heights (h) and interpillar distances (d) have been evaluated, the optimum pillar height being ~70 μm in order to achieve homogeneous lithiation and high cell capacity [24].

The influence of geometry in the performance of conventional and unconventional lithium-ion batteries was studied maintaining the same area of the different components and it has been shown that the geometry with the best performance is the interdigitated structure [25].

Taking into account the state-of-the art on 3D battery architecture simulation and that interdigitated structures maximize the performance of the battery, the goal of this work is focus in the quantitative evaluation of the effect of the variation of the geometrical parameters of the interdigitated structure towards performance optimization of lithium-ion batteries. The considered geometrical parameters are the number, thickness and length of the digits, and the optimization has been performed considering different scan rates. To our knowledge these effects have never been comprehensively reported before and it is important to take them into account before experimentally implementing the adequate geometry of a battery for particular applications, allowing to improve battery design for specific area restrictions. The performance of the battery was determined in two dimensions at different scan rates up to 400 °C, as the combination of interdigitated structure fabrication with printing technologies allows to obtain interdigitated batteries with small size and thickness and yet with high delivered capacity. The optimization of the interdigitated structure by a FEA was carried out taking into account the number, thickness and length of the digits, while maintaining the area of the different components constant. The results are also compared with a conventional structure. As a result, optimization of the geometrical parameters of interdigitated geometries is achieved, allowing to guide experimental fabrication by providing an essential tool for proper battery design and implementation.

## 2. Theoretical simulation model and parameters

The main components of lithium ion batteries are the anode, the cathode and the separator, that can be simulated by the Doyle/Fuller/Newman model in two dimensions (2D) [26].

Depending on the boundary conditions, the main equations governing the performance of the electrodes (anode and cathode) are (see Annex I for the nomenclature and the physical meaning of the different symbols):

- diffusion and ionic conductivity of lithium ions in electrolyte and electrodes:

$$\varepsilon_{a,c}\frac{\partial C_L}{\partial t}=\frac{D_{eff,c,a}}{\delta^2}\frac{\partial^2 C_L}{\partial x^2}+a\left(1-t_+^0\right) \tag{1}$$

- relation between the potential of the electrolyte and the local current density in the electrodes (Ohm's law):

$$-\frac{K_{eff,c,a}}{\delta^2}\frac{\partial^2\phi_L}{\partial x^2}=Faj_{Li+}+\frac{2kRT}{F}\left(1-t_+^0\right)\frac{\partial^2 \ln C_L}{\partial x^2} \tag{2}$$

$$-\frac{\sigma_{eff,c,a}}{\delta^2}\frac{\partial^2\phi_E}{\partial x^2}=-FaJ_{Li+} \tag{3}$$

- the diffusion of lithium ions in the active material:

$$\frac{\partial C_E}{\partial t}=D_{Li}\left[\frac{\partial^2 C_E}{\partial r^2}+\frac{2}{r}\frac{\partial C_E}{\partial r}\right] \tag{4}$$

- conductivity and diffusion:

$$\sigma_{eff,c,a}=\sigma_{c,a}\left(1-\varepsilon_{c,a}-\varepsilon_{f,c,a}\right) \tag{5}$$

$$D_{eff,c,a}=D_l\varepsilon_{c,a}^{brugg},\ brugg=1.5 \tag{6}$$

The relevant equations for battery separators are:

- Lithium-ion diffusion in the electrolyte:

$$\varepsilon_s\frac{\partial C_L}{\partial t}=\frac{D_{eff,s}}{\delta^2}\frac{\partial^2 C_L}{\partial x^2} \tag{7}$$

- Electrical conductivity:

$$\kappa_f=\kappa_l\frac{\varepsilon_s}{\tau^2} \tag{8}$$

In this work, a finite element method is implemented, considering the electrochemical and transport processes in interdigitated lithium ion battery structure such as: [porous positive electrode, ($Li_xMn_2O_4$) | porous separator, poly(vinylidene-trifluoroethylene) (P(VDF-TrFE))