

Design of scalable three-dimensional electrode and device architectures with good rate capabilities

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3-D architectures are electrode and battery configurations that are non-planar configurations and offer the potential for greatly improved performance, particularly in terms of volumetric energy density, compared to traditional planar (1-D along the cell-sandwich and 2-D along the plane of the electrodes) configurations (or their derivatives).<sup>1,2</sup> In this work, we present a novel 3-D electrode and device architecture that can potentially provide significant enhancements in energy density. These designs are expected to be scalable and to operate over length scales that exhibit rapid electron and ion transport.

One of the electrode designs we have explored is shown in Fig. 1 and is based on a photonic crystal design.<sup>3</sup> The local current collector framework arranged in a 3-D configuration is shown in the bottom right-hand corner of Fig. 1a. We refer to this structure as a 3-D scaffold and these structures can be fabricated from carbon-based or metallic materials. The scaffold is coated conformally with a redox-active electrode material. The resulting porous 3-D electrode is shown in the cut-away sections of Fig. 1a and the entire electrode is shown more clearly in Fig. 1b. The continuous pore volume (Fig. 1b) is a key element for this electrode and is purposely designed to not only be interconnected but also to be minimized. Liquid electrolyte, which fills the pores, will thus have access to all of the redox-active material at a suitable length scale. A detailed view of how the electrolyte is arranged within a given electrode element is shown in Fig. 1c. This figure also indicates how the electrolyte is continuous through the well-structured electrode architecture leading to very low tortuosity.

The development of non-planar architectures has far reaching effects because, as our calculations show, it will be possible to achieve a significant increase in volumetric energy density without relying on the development of new materials. Current batteries are 2-D devices and, effectively, so are the electrodes that constitute the battery. Research has shown that electrodes which provide electrochemical reactions throughout a 3-D

volume lead to better utilization of electrode materials (increased energy density) and higher power density because of short diffusion lengths which minimize ohmic loss. However, transitioning from a 3-D material to a 3-D electrode design has proven to be problematic. A few researchers have demonstrated 3D electrode architectures<sup>4</sup> at a nanoscale level. In such cases, the normalized gravimetric densities are very good, but the total amounts of energy and power are low because of the small amount of material being used. In this paper, we will present design strategies that effectively scale-up the nanoscale features so that electrodes at millimeter and larger dimensions effectively possess the same energy and power densities that are exhibited at the nanoscale. We will also present different electrode material systems with 75 vol% active material that are able to provide volumetric energy densities that are 3 to 5 times higher than that of planar electrodes. This significant improvement is possible due to high vol% of the active material and the ability to pack the active material at nearly the crystal density rather than ‘tap density.’ We still retain good power characteristics because of a well-connected 3-D transport network with tortuosity close to 1.0. Possible 3-D cell configurations that demonstrate excellent energy densities at the cell-level will also be discussed.

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