Three-dimensional Nanostructuring of Metal-Organic Frameworks for Energy Applications

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Metal-organic frameworks (MOFs) are a growing family of microporous materials that are attracting significant research and industrial interest, thanks to their unique features including a tuneable and narrow intrinsic pore size distribution, and very high surface area. MOFs are also being increasingly used as a precursor for the preparation of well-dispersed metal-containing carbon electrodes. A major challenge remains the three-dimensional structuring of MOFs into hierarchical functional morphologies and their integration as active elements in electrical material systems, due to their usually low electronic conductivities. Here, we present some flexible strategies for the structuring of MOFs into three-dimensional morphologies and their application for energy storage. In particular, we focus on lithium–sulfur (Li–S) batteries as a promising system for the next-generation of high energy density batteries. Despite progress, the energy density of current Li–S technologies is still below that of conventional intercalation-type cathode materials, due to limited stability and utilization efficiency at high sulfur loading.

We demonstrate a highly performing free-standing monolithic electrode architecture for Li–S batteries with superior electrochemical stability and energy density [1]. The electrode layout consists of a highly conductive three-dimensional network of N,P co-doped carbon with well-dispersed metal-organic frameworks nano-domains of ZIF-67 and HKUST-1. The hierarchical monolithic 3D carbon networks provides an excellent environment for charge and electrolyte transport as well as mechanical and chemical stability. The electrically-integrated MOF nano-domains significantly enhance the sulfur loading and retention capabilities by inhibiting the release of lithium polysulfide specifies as well as improving the charge transfer efficiency at the electrolyte interface. Our optimal 3D carbon-HKUST-1 electrode architecture achieves a very high areal capacity of > 16 mAh cm^{-2} with a capacity retention of 82 % at 0.2C for over 300 cycles, providing an attractive candidate material for future high energy density Li–S batteries. We believe that our approaches provide some promising directions for the engineering of functional MOF morphologies for electrochemistry.

References

1. Liu, B.; Bo, R.; Taheri, M.; Di Bernardo, I.; Motta, N.; Chen, H.; Tsuzuki, T.; Yu, G.; Tricoli, A. *Nano Lett.* **2019,** 19, (7), 4391-4399.