Accelerated Discovery of Next-generation Battery Materials using Atomic-scale Computer Simulations

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Rechargeable batteries play a crucial role in the transition to a clean energy infrastructure, but until now, the development of novel, low-cost, high capacity and/or high voltage electrode materials, has been slow. The performance and durability of existing batteries has been hampered by our limited understanding of the complex processes taking place inside the electrode and electrolyte materials and at the battery interfaces, e.g. at the solid electrolyte interphase (SEI) at the negative electrode and the cathode electrolyte interphase (CEI). Despite decades of research, the fundamental reaction, degradation and transport mechanisms have remained elusive for a large number of potentially promising battery materials, and the development of new, high performance materials therefore hinges on an improved understanding of these mechanisms. Similarly, the development of next-generation battery technologies, e.g. metal-sulfur and metal-air batteries, has been limited by the lack of a detailed understanding of the fundamental reaction mechanism which limits the efficiency, accessible capacity and durability of these batteries [1].

Atomic-scale computer simulations at the level of density functional theory (DFT) calculations and beyond have now reached predictive accuracy in many critical areas of materials design and characterization, enabling accelerated discovery of novel, high-performance battery materials and identification of rate- and potential limiting reaction steps. Here, we will provide a number of recent examples of how such simulations can been used efficiently to identify, e.g., the limiting ionic and electronic transport mechanisms in novel Li-ion electrode materials like $LiFeBO_3$, with [2] or without protective coatings [3], and in Li-S batteries [4]. We will also show why differences in thermodynamics and kinetic overpotentials between Li-O_2 and Na-O_2 batteries can lead to large differences in observed discharge products [5], and how dopants and additives can be used to improve the performance of negative [6] and positive electrode materials and catalysts in secondary Zn-O_2 batteries [7,8].

Finally, we will show how machine learning and cluster expansion techniques can be used to accelerate the rate of discovery of new electrode materials even further.

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