

# Interfacial chemistry and electrolyte approaches for next generation batteries

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Continuous solid electrolyte interphase (SEI) and dendrite growth, as well as formation of ion blocking interfaces are some of the crucial issues preventing the commercialization of batteries depending on the implementation of Li/Na/K/Mg/Ca metal anodes.<sup>1,2,3</sup> In the first part of my talk, I will focus on the chemistry of formed interfaces, including growth and ion transport control. Second part of my talk deals with bulk liquid, polymer and hybrid electrolyte molecular structure approaches for circumventing dendrite formation.

Understanding of the SEI growth on alkali and alkaline earth metals in contact with liquid electrolytes under open circuit potential conditions important for shelf aging is still in its infancy. I will show that the SEI formed on Li in contact with glyme-based electrolyte grows via reaction-controlled mechanism that is rapidly substituted by the diffusion-controlled one.<sup>3</sup> The ionic transport and stability of the complex composite SEI material under current is highly dependent on the salt chemistry, with detrimental effects of  $\text{Li}_x\text{S}_y$  and positive effects of the  $\text{Li}_3\text{N}$  constituents. Most recent ion transport and morphological studies show that the initial SEI formed in contact with liquid electrolytes on Li, Na and Mg is nanoporous.<sup>4,5</sup> Based on the electrochemical impedance spectroscopy measurements in symmetric cells and related equivalent circuit models, I will discuss potential growth mechanism via SEI densification. Finally, I will present a possibility of using gas/solid synthesis for preparation of sulfide-based artificial SEIs for both liquid-based and solid-state batteries.<sup>6</sup>

Dendrite formation on alkali and alkaline earth metal anodes is closely related to the ion concentration gradients formed near the electrodes in contact with the electrolytes. I will discuss the method to circumvent this problem by using electrolytes with high cationic transference number. In soft matter electrolytes this is possible when (i) interfacial effect enables preferential anionic adsorption in liquid/solid electrolytes,<sup>7</sup> (ii) the size and steric effects control the association and mobility of the anion in the liquid electrolytes,<sup>8</sup> (iii) ion transport proceeds through percolating amorphous clusters in polymer-in-salt electrolytes.<sup>9</sup>

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