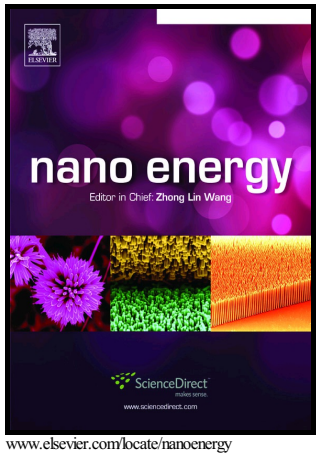

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Abstract:

Safe and high-energy-density lithium rechargeable batteries are urgently required for vehicle electrification and grid energy storage. All-solid-state lithium metal batteries (ASSLMBs) are regarded as a good choice to meet these stringent requirements. However, interfacial instability between Li metal and solid-state sulfide electrolytes (SEs) and lithium dendrite formation are main challenges to be overcome. In this work, molecular layer deposition (MLD) is employed for the first time to develop an inorganic-organic hybrid interlayer (alucone) at the interface between the Li metal and SEs. It is found that the alucone layer can serve as an artificial solid electrolyte interphase (SEI). As a result, interfacial reactions between Li and SEs are significantly suppressed by intrinsically blocking electron transfer at the interface. In addition, lithium dendrites are also suppressed. Coupled with a $LiCoO_2$ cathode, ASSLMBs with 30 MLD cycles of alucone on Li metal exhibit a high initial capacity of 120 mAh g^{-1} and can retain a capacity of 60 mAh g^{-1} after 150 cycles. This work exemplifies the use of MLD to stabilize the interface between SEs and Li metal for ASSLMBs.