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Structure, Chemistry, and Charge Transfer Resistance of the Interface between $Li_7La_3Zr_2O_{12}$ Electrolyte and $LiCoO_2$ Cathode

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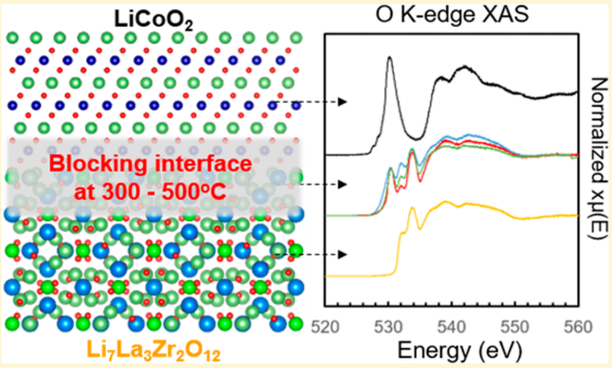
Ⓢ *Supporting Information*



ABSTRACT: All-solid-state batteries promise significant safety and energy density advantages over liquid-electrolyte batteries. The interface between the cathode and the solid electrolyte is an important contributor to charge transfer resistance. Strong bonding of solid oxide electrolytes and cathodes requires sintering at elevated temperatures. Knowledge of the temperature dependence of the composition and charge transfer properties of this interface is important for determining the ideal sintering conditions. To understand the interfacial decomposition processes and their onset temperatures, model systems of $LiCoO_2$ (LCO) thin films deposited on cubic Al-doped $Li_7La_3Zr_2O_{12}$ (LLZO) pellets were studied as a function of temperature using interface-sensitive techniques. X-ray photoelectron spectroscopy, secondary ion mass spectroscopy, and energy-dispersive X-ray spectroscopy data indicated significant cation interdiffusion and structural changes starting at temperatures as low as 300 °C. $La_2Zr_2O_7$ and Li_2CO_3 were identified as decomposition products after annealing at 500 °C by synchrotron X-ray diffraction. X-ray absorption spectroscopy results indicate the presence of also $LaCoO_3$ in addition to $La_2Zr_2O_7$ and Li_2CO_3. On the basis of electrochemical impedance spectroscopy and depth profiling of the Li distribution upon potentiostatic hold experiments on symmetric LCO|LLZO|LCO cells, the interfaces exhibited significantly increased impedance, up to 8 times that of the as-deposited samples after annealing at 500 °C. Our results indicate that lower-temperature processing conditions, shorter annealing time scales, and CO_2-free environments are desirable for obtaining ceramic cathode|electrolyte interfaces that enable fast Li transfer and high capacity.

■ INTRODUCTION

One of the most exciting prospects of solid electrolytes is the possibility of using Li metal as the anode, which would greatly increase the gravimetric energy density over intercalation anodes.[1−6] A number of solid electrolytes with conductivities

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