Supporting information:

Conductivity of Cold Sintered Diphasic Composites Containing a Ceramic Active Material and a Solid-State Electrolyte or Carbon for All Solid-State

Batteries

Zane M. Grady^{1,2}, Zhongming Fan^{1,2}, Julian Fanghanel^{1,2}, Clive A. Randall^{1,2}

¹Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA, United States

²Materials Research Institute, The Pennsylvania State University, University Park, PA, United States



Figure S1 NVP and carbon prior to mixing (A). *NVP* and carbon after mixing (B). *NVP* and carbon pellet after being heated to 510 °C under air (C). *NVP* and carbon after being heated to 637°C under air (D).



Figure S2 The relative density (A) and Archimedes density (B) of carbon black – NVP composites. The relative density (C) and Archimedes density (D) of NZSP – NVP composites. Theoretical densities calculated based on volume fraction of composite and material densities (NVP = 3.16, NZSP = 3.26, carbon black = 1.6 g.cm^{-3})



Figure S3 The logarithm of total, grain boundary (gb), and grain interior (grain) conductivity plotted against inverse absolute temperature for the pure cold sintered NVP.



Figure S4 Complex impedance spectra of a sample containing 2.25 vol.% carbon black. The dispersion in the low frequency data renders it difficult to fit (A), but R_{tot} can be estimated from the well-resolved high frequency semicircle, as shown in (B)



Figure S5 The EIS spectra of all carbon-NVP composites at 30°C. The data for Super P is shown in A and that for carbon nanofibers is shown in B.



Figure S6 Additional TEM images demonstrating carbon presence in the grain boundaries of a carbon black – NVP diphasic composite.



Figure S7 $E_{a,tot}$ is plotted as a function of volume fraction of NZSP (A) or carbon (B)



Figure S8 The EIS spectra of a 90 vol.% NZSP sample used for Mott-Schottky experiments. Multiple temperatures are shown to illustrate the reason for choosing 70°C. The high frequency semicircle is the NZSP response, the intermediate frequency semicircle is the interfacial response. The lines are only meant as a guide to the eye, not an equivalent circuit fitting.