Supporting Information

Decorating β"-alumina solid-state electrolyte with submicron Pb spherical particles for improving Na wettability at lower temperatrues

Hee Jung Chang^a, Xiaochuan Lu^a, Jeffery F. Bonnett^a, Nathan L. Canfield^a, Keesung Han^b, Mark H. Engelhard^c, Keeyoung Jung^{d,*}, Vincent L. Sprenkle^a, and Guosheng Li^{a,*}

^a Electrochemical Materials and Systems Group, Pacific Northwest National Laboratory, 902 Battelle Blvd., Richland, WA 99352, USA

^b Materials Science Group, Pacific Northwest National Laboratory, 902 Battelle Blvd., Richland, WA 99352, USA

^d Interfacial Sci's & Simulation Group, Pacific Northwest National Laboratory, 902 Battelle Blvd., Richland, WA

^d Materials Research Division, Research Institute of Industrial Science & Technology, Pohang, South Korea



Figure S1. SEM images and EDX mapping (oxygen and carbon) for LAT_210°C and LAT_275°C BASEs.



Figure S2. Wetting angles measurements for molten Na drops on BASE surfaces using a sessile drop technique at various temperatures of 150, 175, 200, 225, and 250 °C.



Figure S3. The state-of-charge (SOC) for cells with different LAT treatment. (a) the end-of-charge (EOC), (b) the end-of-discharge (EOD), respectively.



Figure S4. Cross section view of LAT_400°C BASE (a) SEM, and (b) Pb mapping.



Figure S5. High energy resolution XPS spectrum.



Figure S6. Na-Pb binary phase diagram.¹



Figure S7. ²³Na MAS NMR spectra for the samples retrieved from the anode of LAT-400°C. Metallic shift (~1130 ppm)² on ²³Na MAS NMR spectrum obtained from the sample retrieved from the anode of LAT-400°C. It shows that the metallic Na are composed of two main components centered around 1132 and 1135 ppm, which are assigned to be pure Na metal and Na alloys, respectively. Green line is 1132 ppm and red line is 1135 ppm.



Figure S8. XRD measurements for BASEs. (a) BASE from cycled LAT-400°C cell at the discharge state, (b) same BASEs after washing away all residuals with DI water. XRD pattern of Na (in black), Na-Pb alloys (NaPb₃ in magenta), and metallic Pb (in blue), respectively.

- 1. Efanov, A. D., Loginove, N. I., Morozov, A. V., and Mikheyev, A. S. (2008). Investigation of Thermodynamic Properties of Sodium-Lead System. J. Phys.: Conf. Ser. *98*, 032013.
- Gotoh, K., Ishikawa, T., Shimadzu, S., Yabuuchi, N., Komaba, S., Takeda, K., Goto, A., Deguchi, K., Ohki, S., Hashi, K., Shimizu, T., and Ishida, H. (2013). Nmr Study for Electrochemically Inserted Na in Hard Carbon Electrode of Sodium Ion Battery. J Power Sources 225, 137-140.