## **Supplementary Information**

## Bayesian-optimization-guided Experimental Search of NASICON-type Solid Electrolytes for All-solid-state Liion Batteries

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**Supplementary Figure S1.** Performance comparison between multi-objective Bayesian optimization (MOBO) (red line) and non-dominated sorting genetic algorithm II (NSGAII) search (blue line). Optimization targets were the Li-ion conductivity at 30 °C and the relative density for 225 extended compositions by interpolation techniques.



**Supplementary Figure S2.** Performance comparison between multi-objective Bayesian optimization (MOBO) (red line) and random search (black line). Note that two MOBO-type schemes were tested, namely the single- and 3-point MOBO as represented by the solid or hatched line, in which experimental evaluation(s) for one or 3 composition(s) is (are) performed in each MOBO iteration, respectively. Optimization targets were the Li-ion conductivity at 30 °C and the relative density. Panels (a) and (b) represent the optimizations for 47 measured and 225 extended compositions by interpolation techniques, respectively.