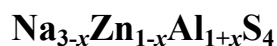


## Supporting Information

### Preparation and characterization of the new solid electrolytes



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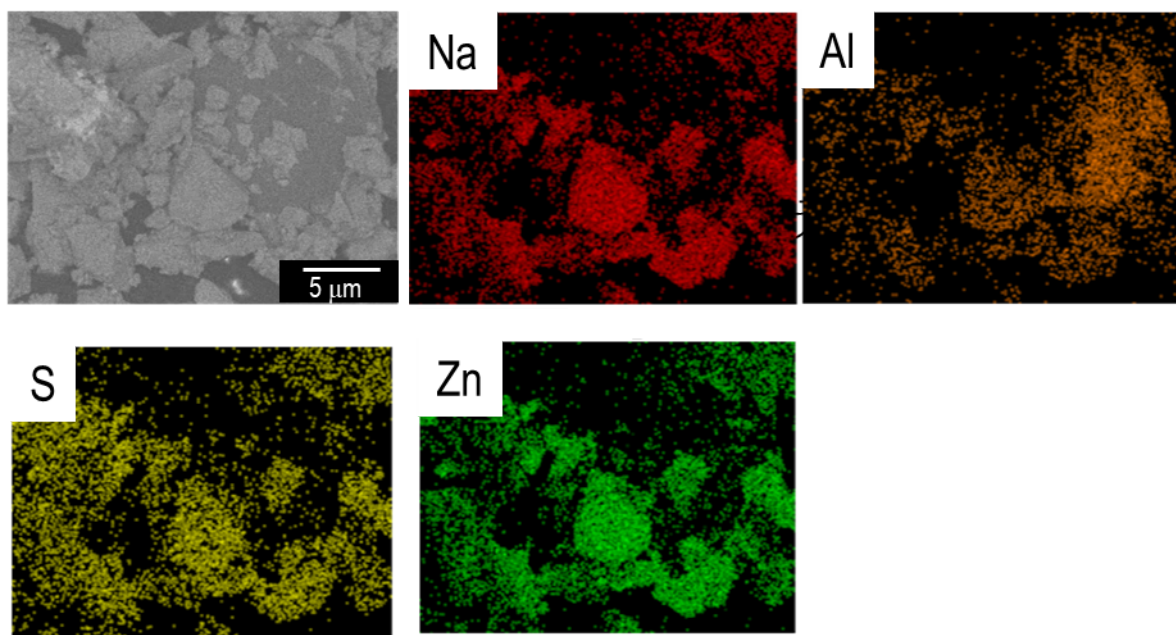
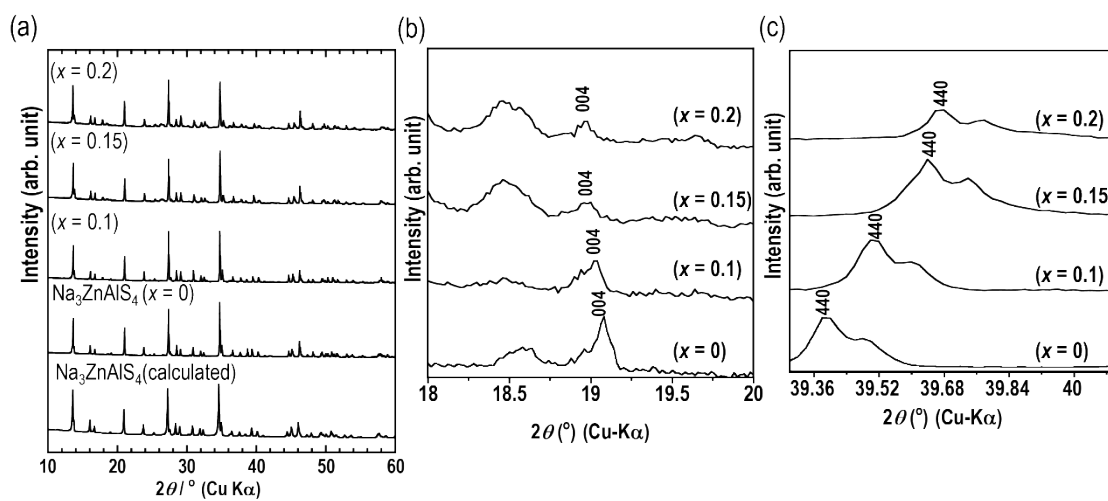


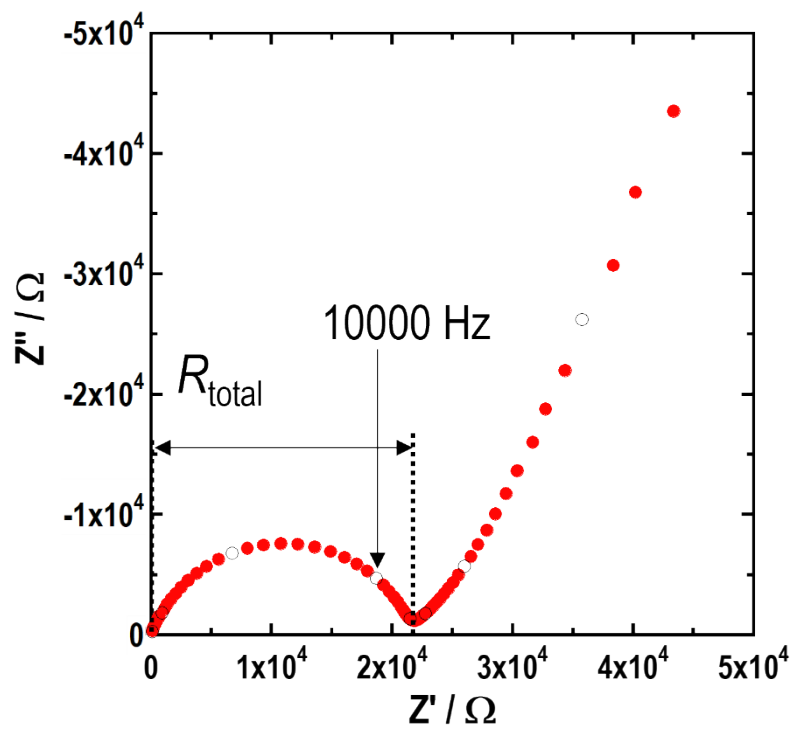
Figure S1. SEM-EDS mappings of  $\text{Na}_3\text{ZnAlS}_4$  electrolyte.

**Table S1.** The unit cell parameters  $a$ ,  $c$ , and  $V$  of  $\text{Na}_{3-x}\text{Zn}_{1-x}\text{Al}_{1+x}\text{S}_4$ , obtained by the Le Bail fitting method.

$x$	$a$ (Å)	$c$ (Å)	$V$ (Å <sup>3</sup> )
0	12.9206(1)	18.5822(2)	3102.12(4)
0.1	12.8942(1)	18.6374(2)	3098.66(6)
0.15	12.8499(3)	18.6787(5)	3084.25(12)
0.2	12.8408(3)	18.6974(6)	3082.93(14)



**Figure S2.** (a) XRD patterns of  $\text{Na}_{3-x}\text{Zn}_{1-x}\text{M}_{1+x}\text{S}_4$  ( $x = 0, 0.1, 0.15, 0.2$ ), (b) Zoom on the Bragg peak 004, and (c) Zoom on the Bragg peak 440.



**Figure S3.** Nyquist plot of  $\text{Na}_{2.9}\text{Zn}_{0.9}\text{Al}_{1.1}\text{S}_4$  at  $29.8 \text{ }^\circ\text{C}$ .