Supporting Information

Uncovering the Structural Evolution in Na-Excess Layered Cathodes for Rational Use of an Anionic Redox Reaction

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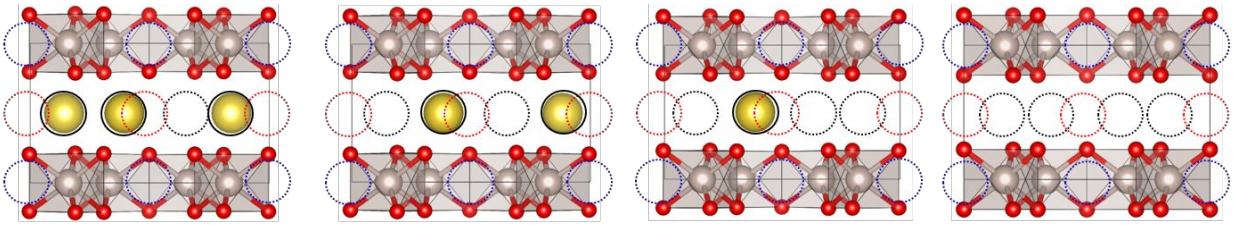

x = 0.625 in $Na_{1-x}Ru_{0.5}O_{1.5}$ x = 0.75 in $Na_{1-x}Ru_{0.5}O_{1.5}$ x = 0.875 in $Na_{1-x}Ru_{0.5}O_{1.5}$ x = 1.0 in $Na_{1-x}Ru_{0.5}O_{1.5}$

Figure S1. Atomic structures in the lowest-energy configurations of $x = 0.625$, 0.75, 0.875, and 1.0 in normalized $Na_{1-x}Ru_{0.5}O_{1.5}$, shown as open circles (2b: blue, 2c: red and 4h: black).

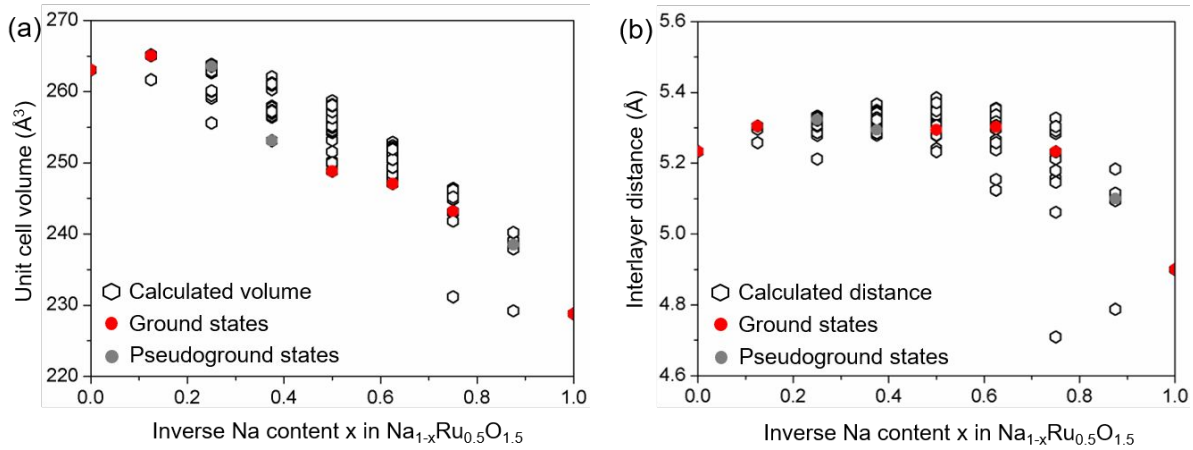

Figure S2. (a) Unit-cell volumes and (b) distances between the $Na_{1/3}Ru_{2/3}O_2$ layers, as functions of inverse Na content ($x = 0.0$ to $x = 1.0$) in normalized $Na_{1-x}Ru_{0.5}O_{1.5}$.

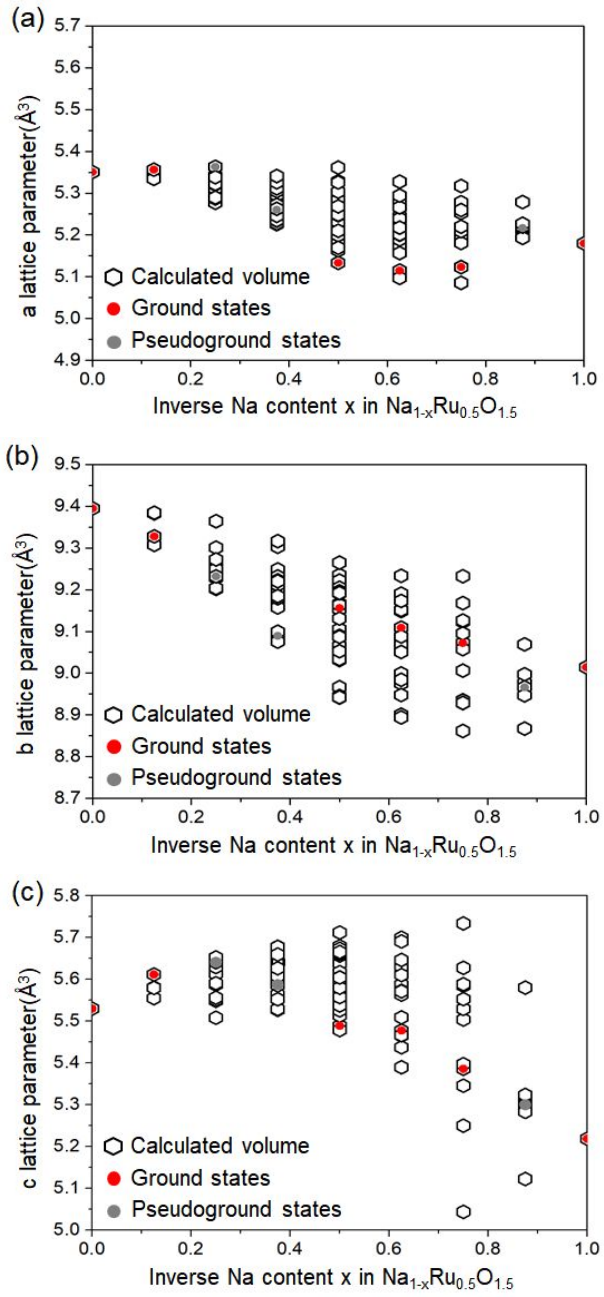

Figure S3. (a) a lattice parameters (b) b lattice parameters (c) c lattice parameters, as functions of inverse Na content ($x = 0.0$ to $x = 1.0$) in normalized $Na_{1-x}Ru_{0.5}O_{1.5}$

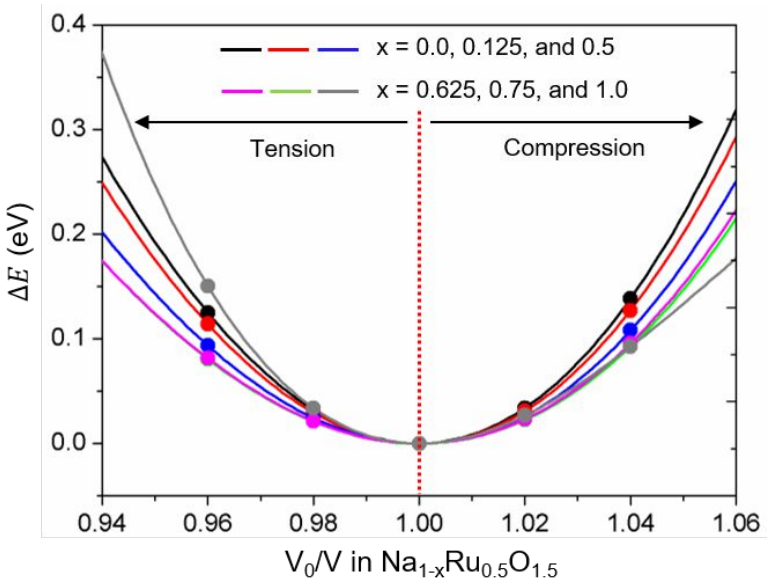

Figure S4. Changes in strain energy as the volume ratio (V_0/V) is varied in $Na_{1-x}Ru_{0.5}O_{1.5}$ with $x = 0.0$, 0.125, 0.5, 0.625, 0.75, and 1.0. The solid lines were fitted by the Birch–Murnaghan equation of states, derived from the total energies by first-principles calculations.

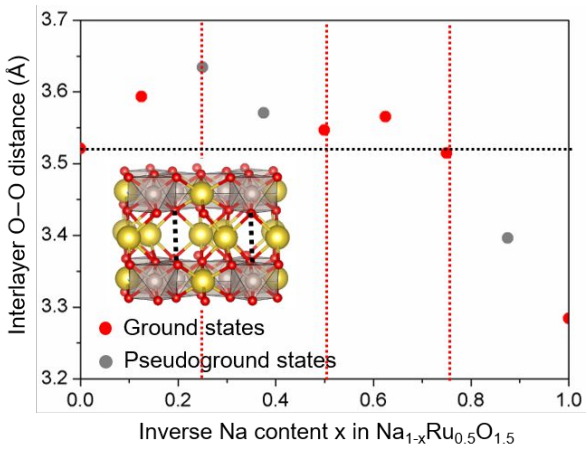

Figure S5. Interlayer O – O bond distance, illustrated as the black dotted line in the inset figure, between the transition metal layers as functions of inverse Na content ($0 \leq x \leq 1.0$) in normalized $Na_{1-x}Ru_{0.5}O_{1.5}$.