# Interplay between A-site and oxygen-vacancy ordering, and mixed electron/oxide-ion conductivity in $n = 1$ Ruddlesden−Popper perovskite $Sr_2Nd_2Zn_2O_7$

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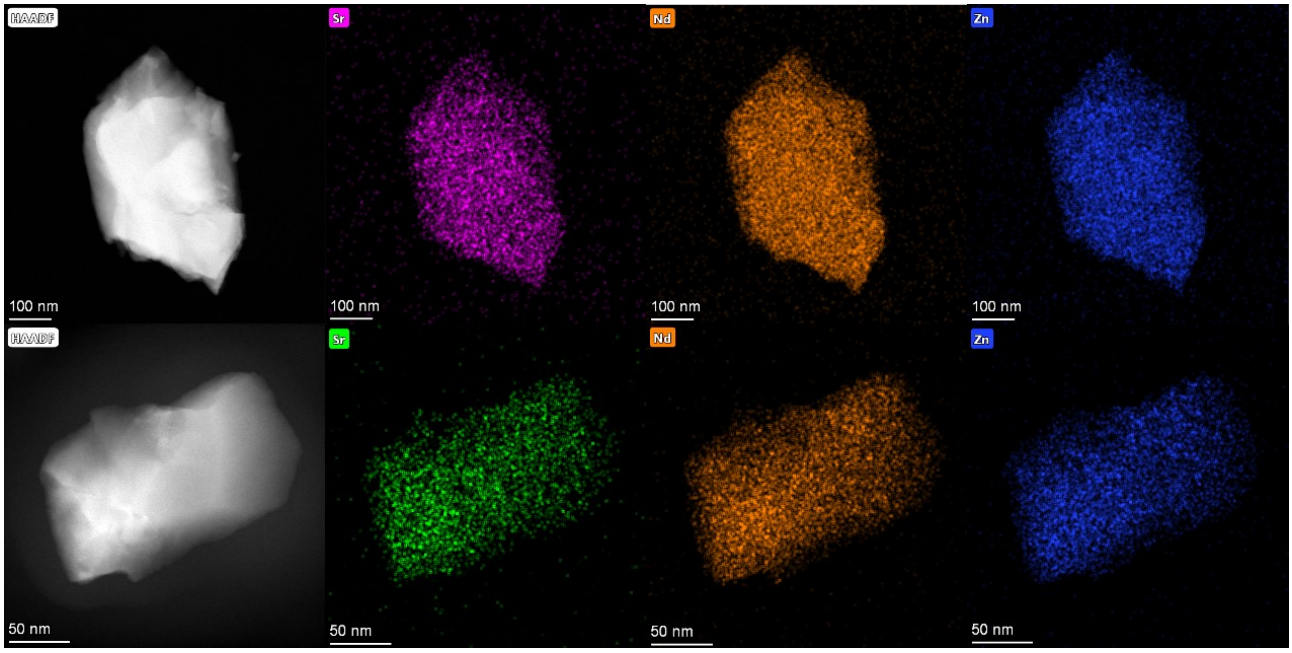


**Fig. S1** TEM maps and elemental mapping images for $Sr_2Nd_2Zn_2O_7$.

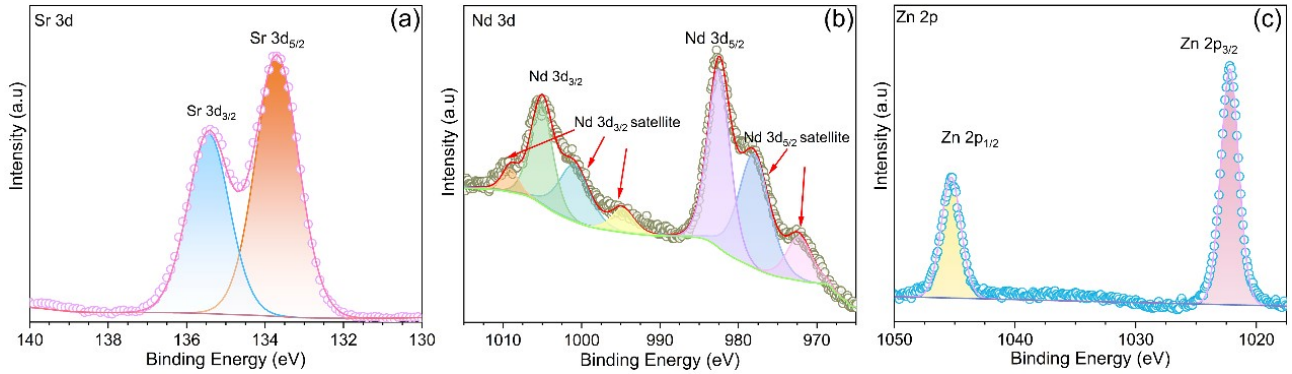


**Fig. S2** High-resolution XPS spectra for Sr 3d (a), Nd 3d (b), and Zn 2p (c)

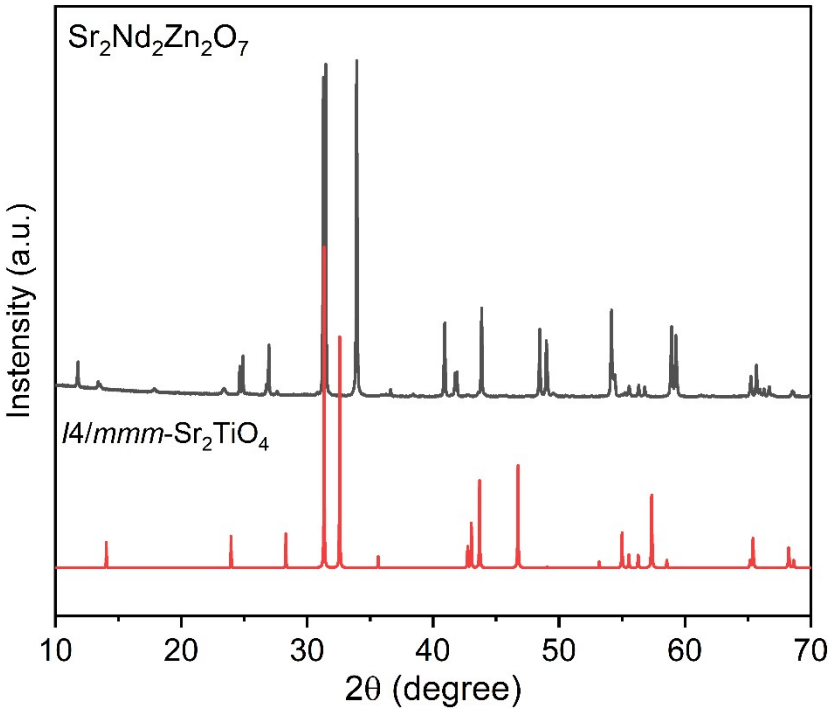


**Fig. S3** The PXRD pattern for $Sr_2Nd_2Zn_2O_7$. A simulated PXRD pattern of *I*4/*mmm*-$Sr_2TiO_4$ is given in the pattern for comparison.

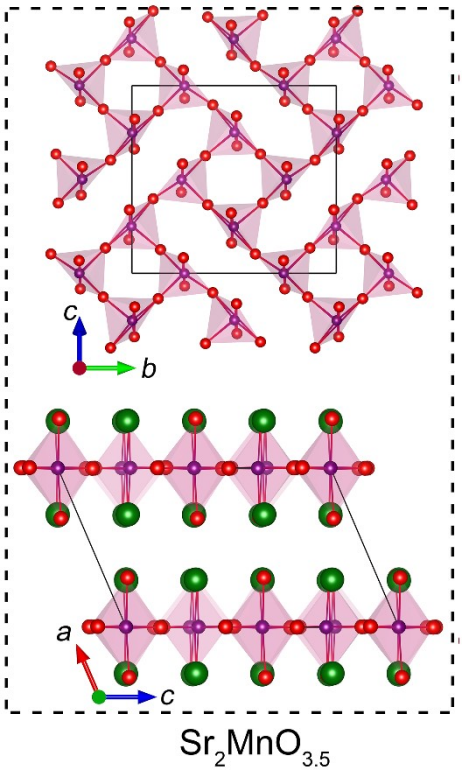


**Fig. S4** Crystal structure of oxygen-vacancy ordered $Sr_2MnO_{3.5}$.

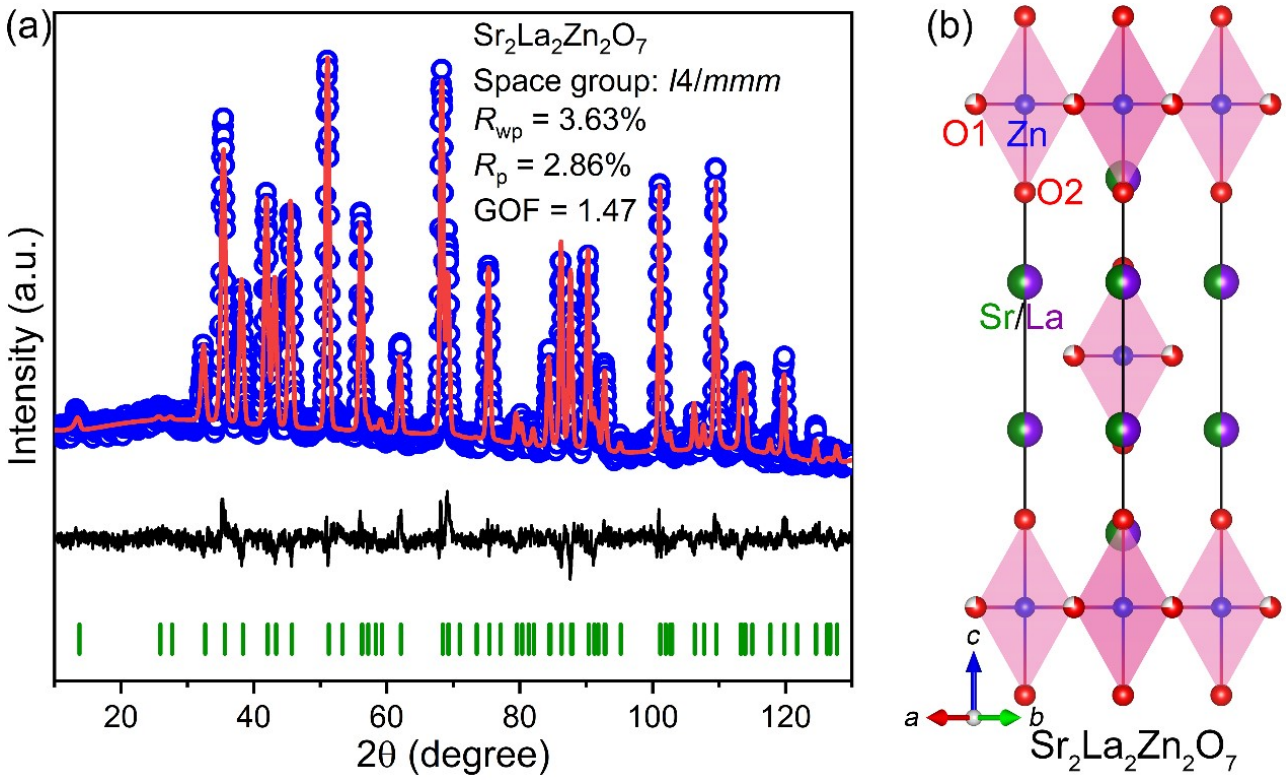


**Fig. S5** (a) Rietveld refinement plots of high-resolution NPD data for $Sr_2La_2Zn_2O_7$. (b) Crystal structure of $Sr_2La_2Zn_2O_7$ viewed along the [110] direction.

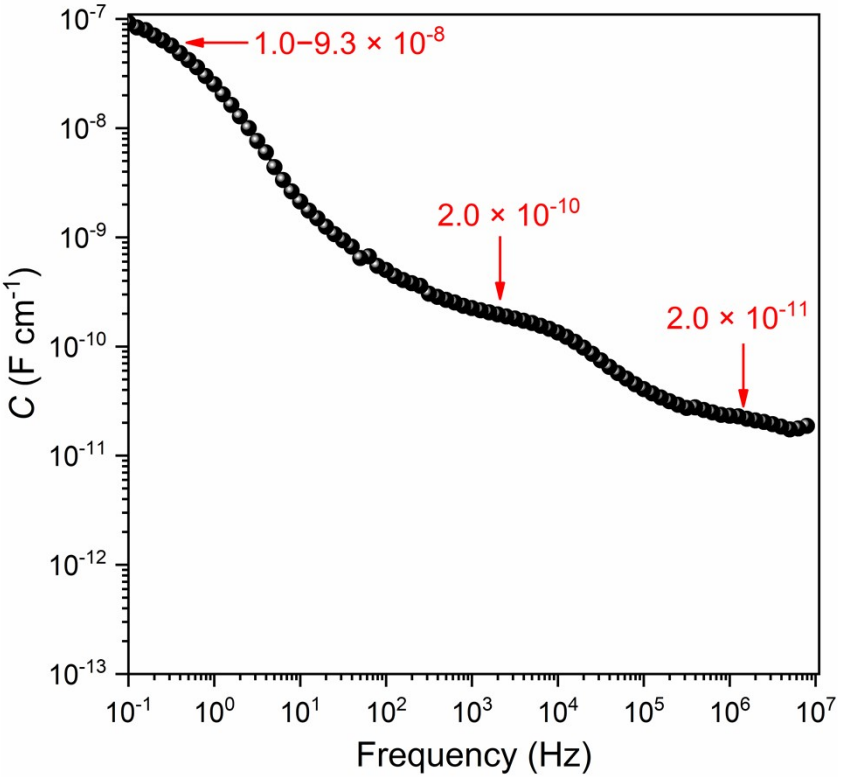


**Fig. S6** Plots of frequency as a function of frequency. The corresponding impedance data were recorded at 550 °C under dry air conditions.

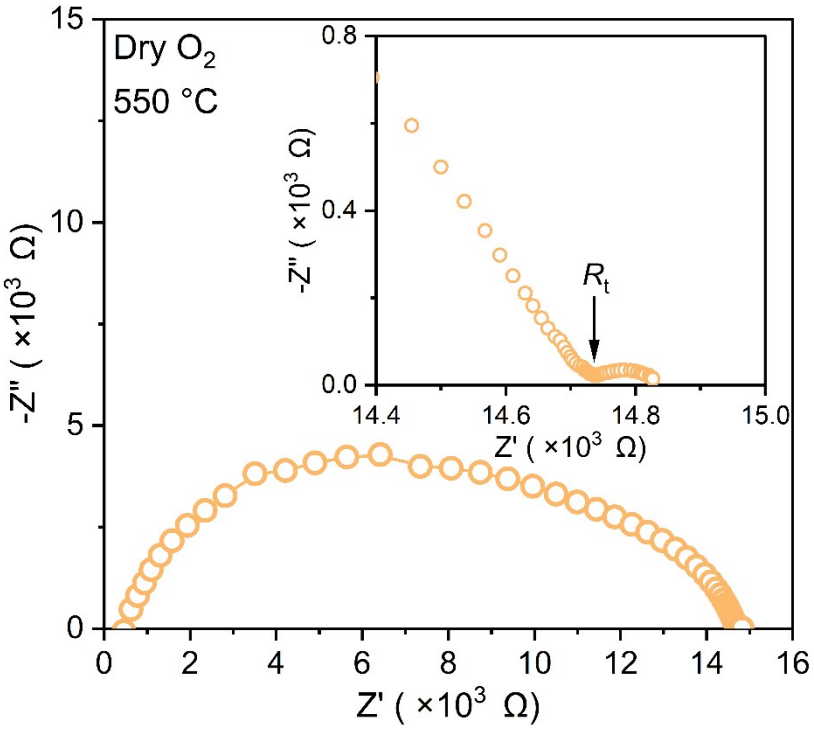


**Fig. S7** Complex impedance spectra of $Sr_2La_2Zn_2O_7$ recorded at 550 °C under $O_2$ atmospheres.

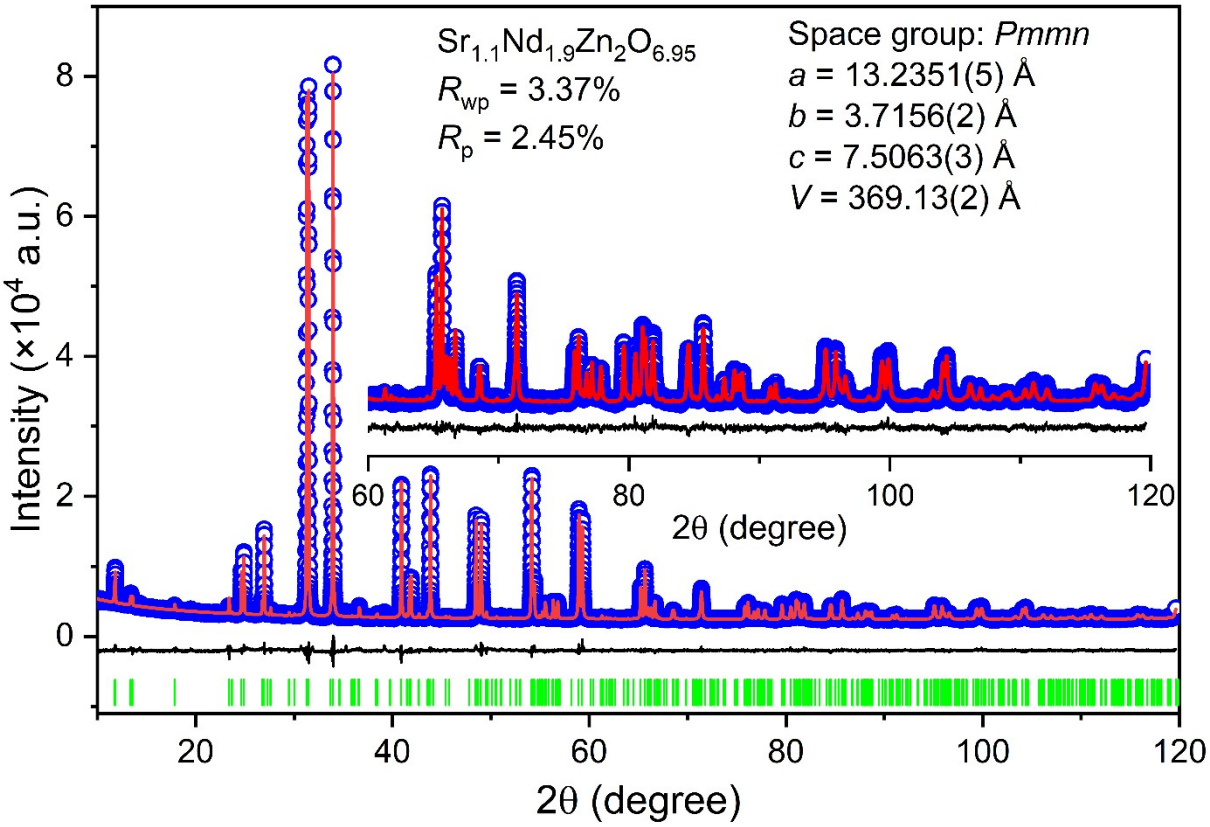


**Fig. S8** Rietveld refinement plot of PXRD data for $Sr_{2.1}Nd_{1.9}Zn_2O_{6.95}$.

**Table S1.** Selected Interatomic bond lengths in $Sr_2Nd_2Zn_2O_7$.

| bond | length (Å) | bond | length (Å) | bond | length (Å) |
|---|---|---|---|---|---|
| Sr1−O5 | 2.418(7) | Nd1−O4 | 2.392(6) | Zn1−O1 × 2 | 1.922(6) |
| Sr1−O2 × 2 | 2.6422(7) | Nd1−O1 | 2.403(6) | Zn1−O2 | 1.96(2) |
| Sr1−O4 × 2 | 2.820(5) | Nd1−O3 | 2.432(5) | Zn1−O4 × 2 | 2.258(6) |
| Sr1−O5 × 2 | 2.861(6) | Nd1−O4 × 2 | 2.498(4) | <Zn1−O> | 2.064(9) |
| Sr1−O3 | 3.022(7) | Nd1−O5 × 2 | 2.500(5) | BVS(Zn1) | 1.99 |
| Sr1−O1 | 3.039(9) | <Nd1−O> | 2.461(5) | Zn2−O2 | 1.86(2) |
| <Sr1−O> | 2.792(5) | BVS(Nd1) | 2.59 | Zn2−O3 × 2 | 1.908(5) |
| BVS(Sr1) | 1.59 | | | Zn2−O5 × 2 | 2.334(7) |
| | | | | <Zn2−O> | 2.070(9) |
| | | | | BVS(Zn2) | 2.05 |

**Table S2.** Atomic Coordinates, occupancies, Isotropic Thermal Displacement Factors of $Sr_2La_2Zn_2O_7$ Obtained from Rietveld Refinement against NPD Data.

| atom | site | *x* | *y* | *z* | Occ. | $B_{iso.}$ (Å$^2$) |
|---|---|---|---|---|---|---|
| Sr/La | 4*e* | 0 | 0 | 0.3538(1) | 0.5/0.5 | 0.69(5) |
| Zn1 | 2*a* | 0 | 0 | 0 | 1 | 1.05(8) |
| O1 | 4*c* | 0 | 0.5 | 0 | 0.75 | 2.6(1) |
| O2 | 4*e* | 0 | 0 | 0.1733(2) | 1 | 1.61(6) |

**Table S3.** Selected Interatomic bond lengths in $Sr_2La_2Zn_2O_7$.

| bond | length (Å) | bond | length (Å) |
|---|---|---|---|
| Sr/La−O2 | 2.445(4) | Zn−O1 × 4 | 1.8741(2) |
| Sr/La−O2 × 4 | 2.6755(6) | Zn−O2 × 4 | 2.347(3) |
| Sr/La−O1 × 4 | 2.727(1) | | |