Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.


**Supporting information for:**

# Improvement of electric insulation in dielectric layered perovskite nickelate films via fluorination

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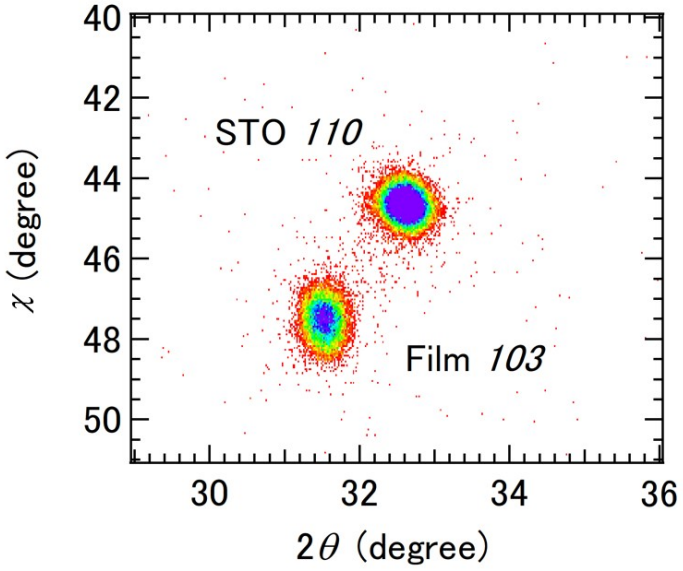



**Figure S1.** Two dimensional XRD $2\theta$–$\chi$ pattern of the $La_{1.5}Sr_{0.5}NiO_4$ film.

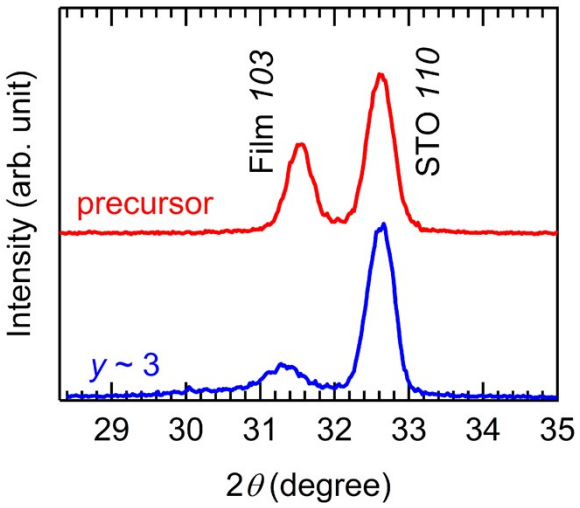
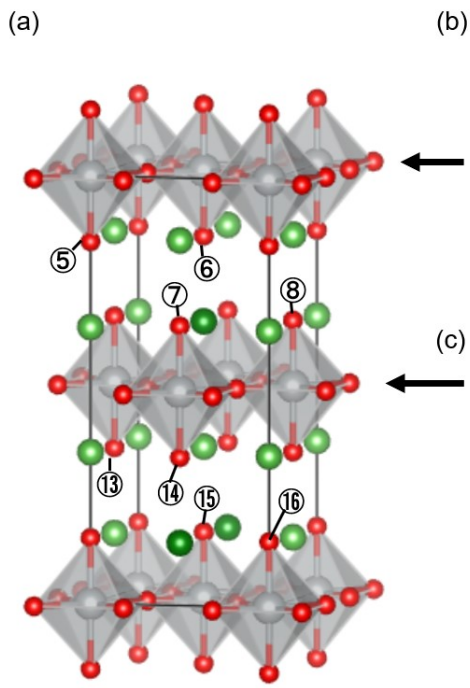



**Figure S2.** $2\theta$–$\omega$ XRD patterns of the precursor and $y \sim 3$ films around the STO *110* and Film *103* diffraction peaks.




(d)

| F position | Total energy (eV) |
|---|---|
| 15, 16 | -191.3394 |
| 5, 16 | -190.6692 |
| 6, 16 | -191.37454 |
| 7, 16 | -191.29407 |
| 14, 16 | -191.44369 |
| 4, 16 | -191.4752 |
| 9, 16 | -191.46278 |
| 5, 6 | -191.33775 |
| 5, 7 | -191.255 |
| 5, 14 | -191.28458 |
| 4, 5 | -191.39563 |
| 5, 9 | -191.50144 |
| 6, 7 | -191.53296 |
| 6, 14 | -191.6956 |
| 4, 6 | -191.54194 |
| 6, 9 | -191.79153 |
| 2, 4 | -190.58805 |
| 1, 4 | -191.12042 |

**Figure S3.** (a) Crystal structure of $La_6Sr_2Ni_4X_{16}$ ($X$ is anion) where La, Sr, Ni and X atoms are drawn as light green, green, gray and red points. (b,c) Top view images of perovskite layers. The number in Fig. (a-c) shows position number of the anions. (d) Fluorine position dependence of total energy of $La_6Sr_2Ni_4O_{14}F_2$. A-site cation configuration was chosen to arrange the Sr atoms as far apart as possible.

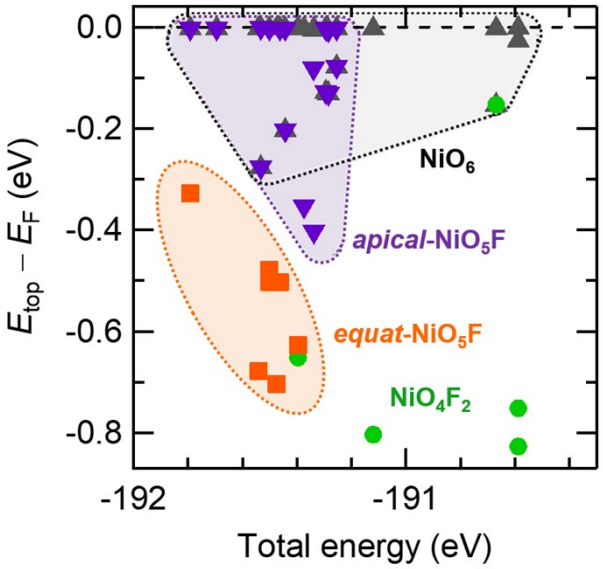



**Figure S4.** The energy level of the top of the Ni 3*d* orbital in the valence band ($E_{top} - E_F$) for the 18 types of $La_{1.5}Sr_{0.5}NiO_{3.5}F_{0.5}$ as a function of total energy. Gray and purple triangles, orange squares and green circles represent $NiO_6$, *apical*- and *equat*-$NiO_5F$ and $NiO_4F_2$ octahedrons, respectively. The models with $NiO_4F_2$ octahedra tended to have higher total energy, indicating that fluorine ions are energetically favored to be located at *apical*- or *equat*-$NiO_5F$. The most stable structure consists of both apical and equatorial configurations. In a structure having only apical configurations, the Ni ions are widely separated, which results in shorter distances between Ni and La/Sr and increases the total energy compared to the structures with both apical and equatorial configurations.

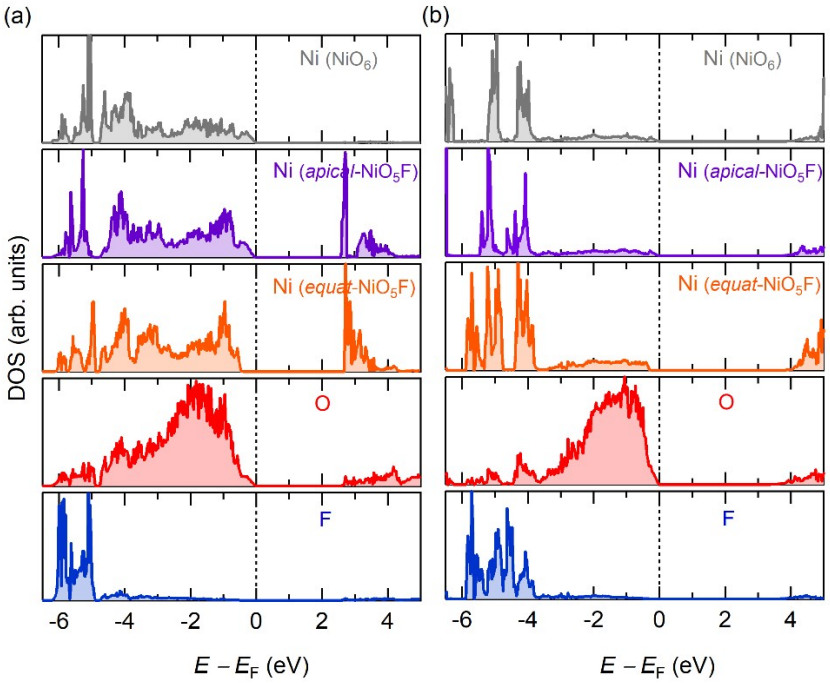



**Figure S5.** Partial DOS for Ni in $NiO_6$, *apical*-$NiO_5F$, and *equat*-$NiO_5F$, O and F sites in $La_{1.5}Sr_{0.5}NiO_{3.5}F_{0.5}$ (Fig. 4(a)) with (a) $U = 4.0$ and (b) 8.0 eV.

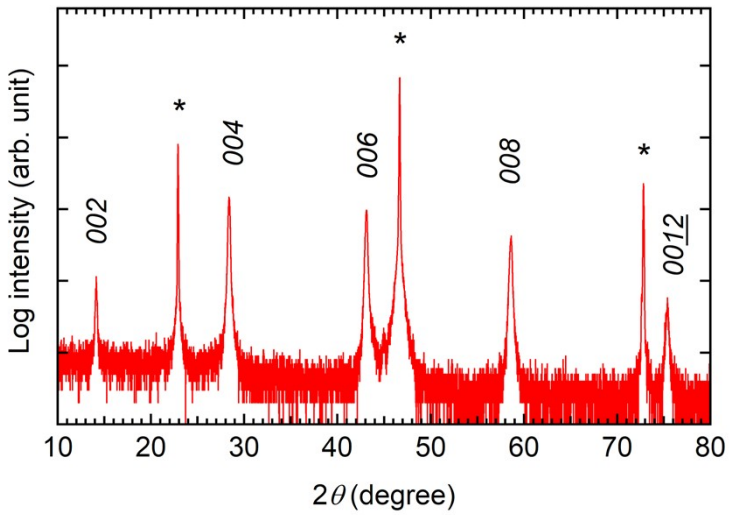



**Figure S6.** Out-of-plane $2\theta$–$\theta$ XRD pattern of the $La_2NiO_4$ film.

**Table S1.** The O and F contents (*x* and *y*) in the $La_{1.5}Sr_{0.5}NiO_xF_y$ films as a function of PVDF treatment condition. The annealing time at 300°C was set to 3 min because the film was broken when the film was annealed with PVDF at 300°C for 24 h.

| PVDF-treated condition | | Chemical composition of $La_{1.5}Sr_{0.5}NiO_xF_y$ films (The *x* and *y* values include error of ~10%) | |
|---|---|---|---|
| temperature (°C) | Time | *x* | *Y* |
| 150 | 10 h | 3.4±0.4 | 0.4±0.1 |
| 200 | 7 h | 3.2±0.4 | 1.0±0.1 |
| 250 | 5 h | 1.9±0.2 | 3.2±0.4 |
| 300 | 3 min | 2.5±0.3 | 2.1±0.3 |