Supplementary Information for

Lanthanum doping enabling high drain current modulation in a p-type tin monoxide thin-film transistor

Sungyeon Yim[1], Taikyu Kim[1], Baekeun Yu[1], Xu Hongwei[1], Yong Youn[2], Seungwu Han[2], and Jae Kyeong Jeong[1]

[1]Department of Electronic Engineering, Hanyang University, Seoul 04763, South Korea

[2]Department of Materials Science and Engineering, Seoul National University, Seoul 08826, Korea

AUTHOR EMAIL ADDRESS: J. K. Jeong (jkjeong1@hanyang.ac.kr)

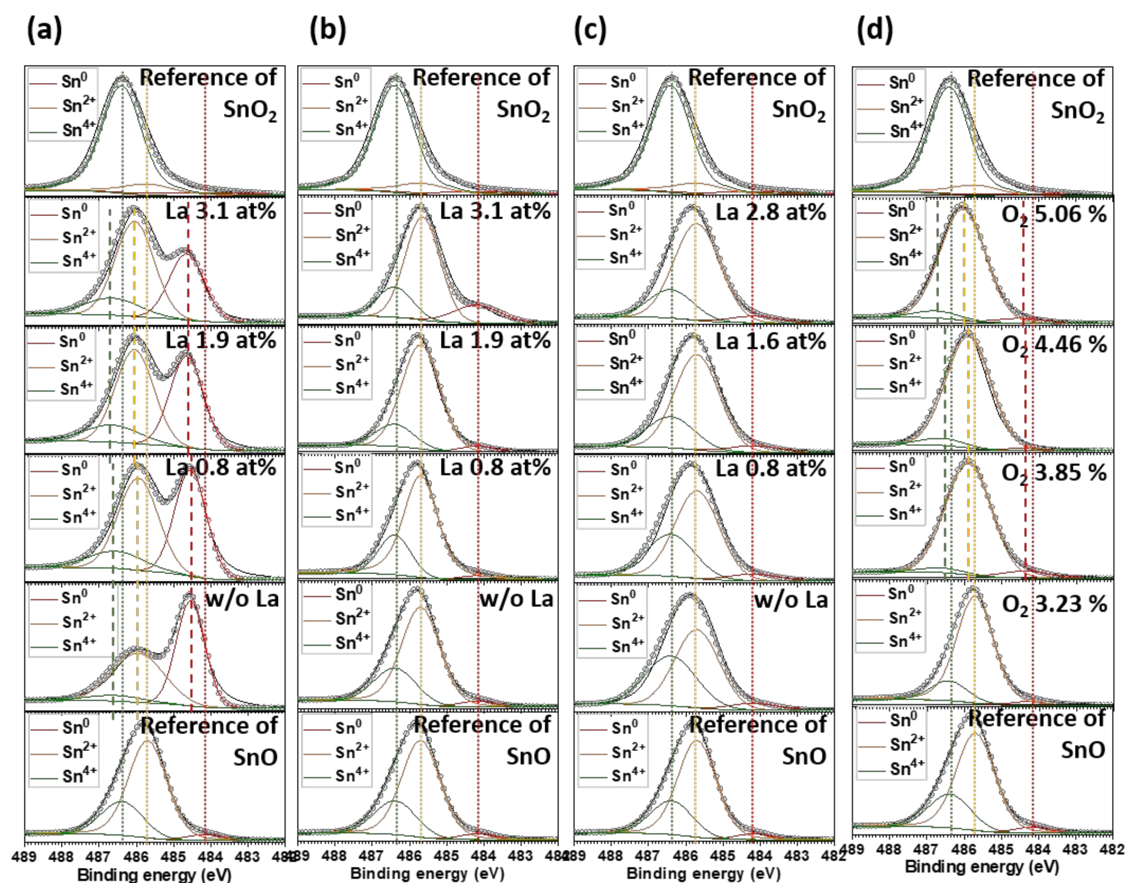

Figure S1. XP spectra of Sn $3d_{5/2}$ for the SnO films with different La loadings (a) before PDA, (b) after PDA at 250 ºC, and (c) after PDA at 300 ºC. (d) XP spectra of Sn $3d_{5/2}$ for the SnO films with different oxygen pressure on 1.9 at% loadings after PDA at 250 ºC.

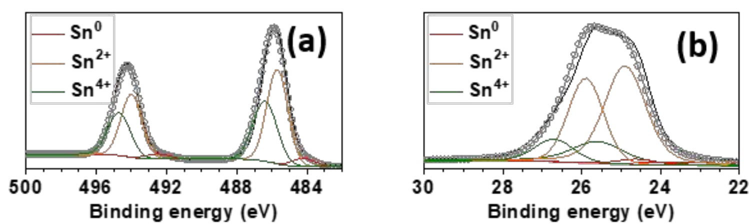

Figure S2. XP spectra of (a) Sn 3d and (b) Sn 4d for the La-free SnO films post-annealed at 250 ºC for 1 hr.

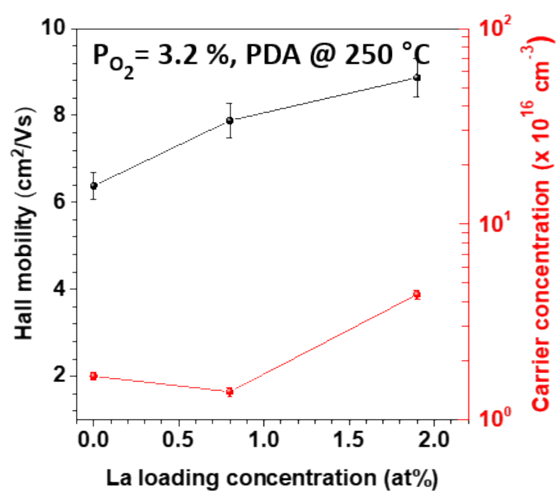

Figure S3. Electrical properties of 250 °C annealed SnO films with different La loadings from Hall effect measurement. Black line (left y-axis) is Hall mobility, and red line (right y-axis) is a free hole carrier concentration measured from Hall measurement.

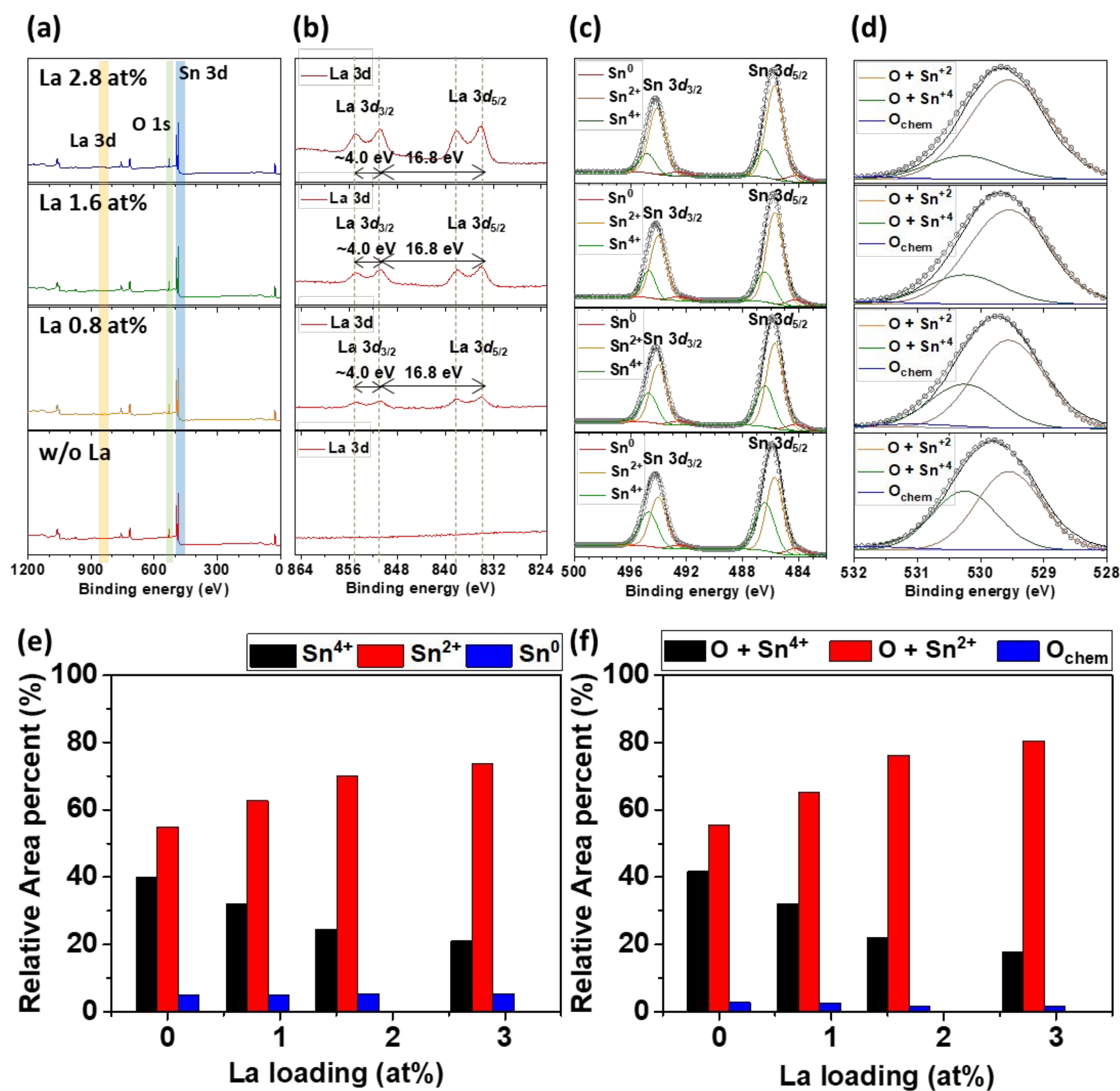

Figure S4. (a) XPS survey spectra, XP spectra of (b) La 3d, (c) Sn 3d, and (d) O 1s for the SnO films annealed in 300 °C with different La loadings. Chemical compositions including Sn^0, Sn^{2+}, and Sn^{4+} of SnO films with different La loadings, which were de-convoluted from XP spectra of (e) Sn 3d and (f) O 1s.

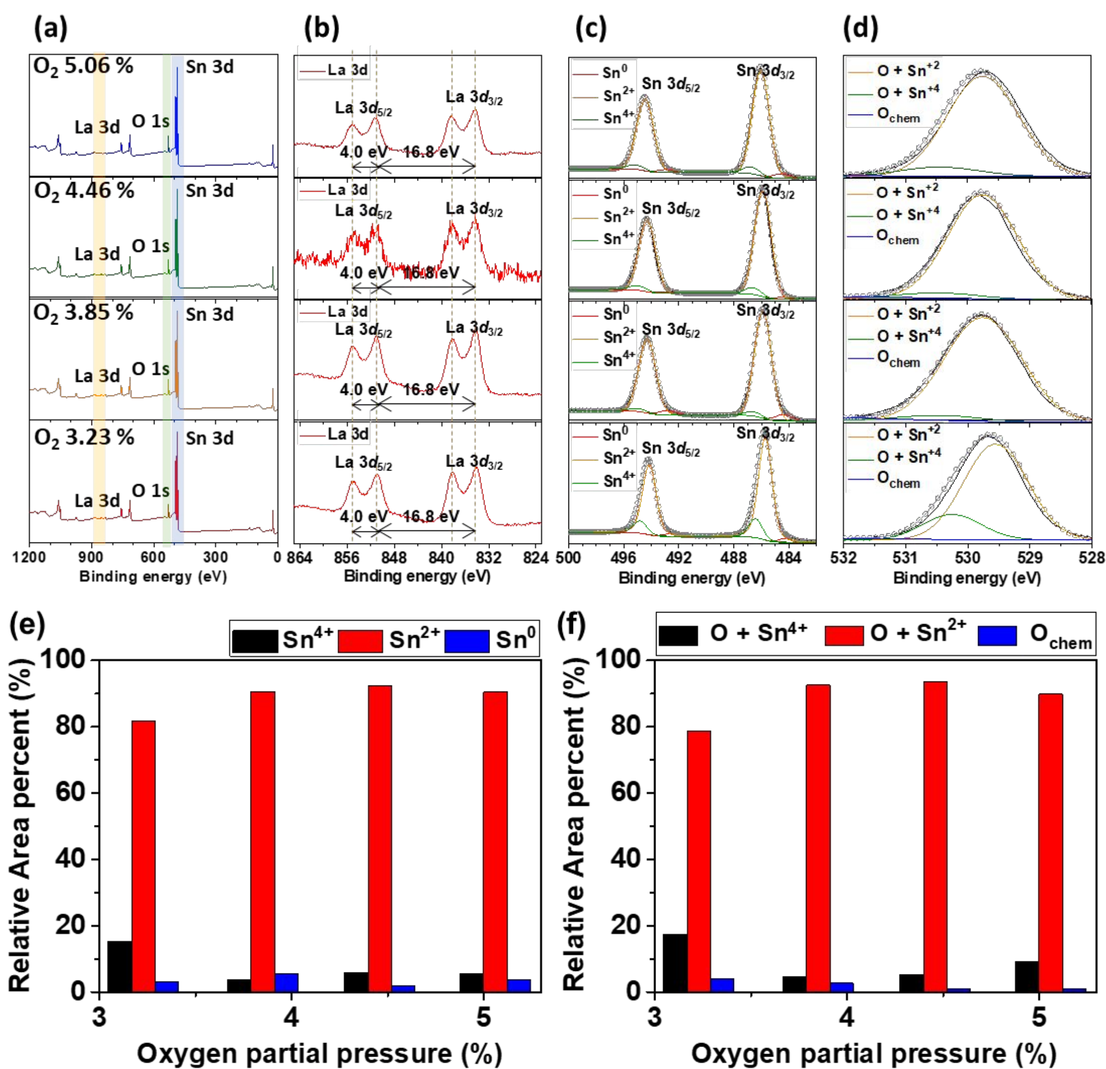

Figure S5. (a) XPS survey spectra, XP spectra of (b) La 3d, (c) Sn 3d, and (d) O 1s for the SnO films annealed at 250 °C with different oxygen pressure on 1.9 at% loadings. Chemical compositions including Sn^0, Sn^{2+}, and Sn^{4+} of SnO films with different oxygen pressure on 1.9 at% La loadings, which were de-convoluted from XP spectra of (e) Sn 3d and (f) O 1s.

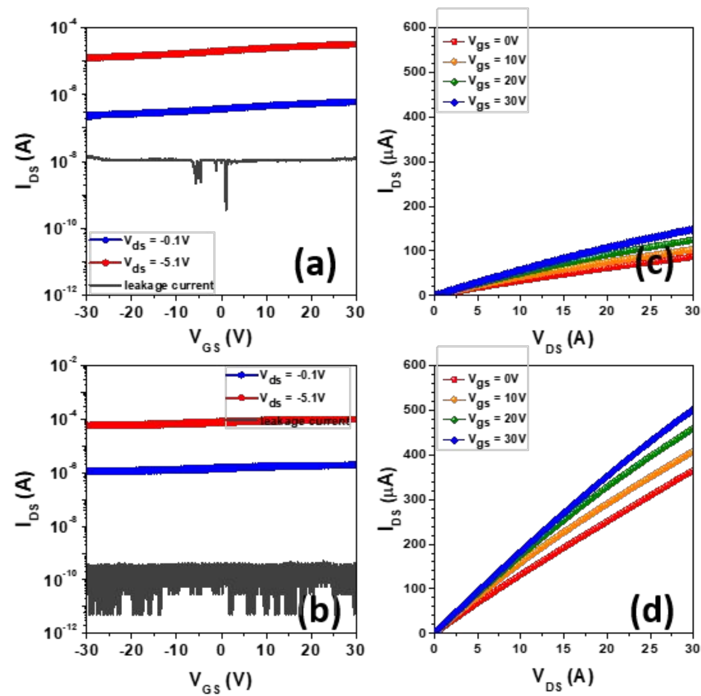

Figure S6. Transfer characteristics of the SnO TFTs with different La loadings of (a) 0 at% and (b) 1.9 at% after the PDA at 250 °C for 1 hr under the forming gas atmosphere. The corresponding output characteristics of the SnO TFTs with different La loadings of (c) 0 at% and (d) 1.9 at%.

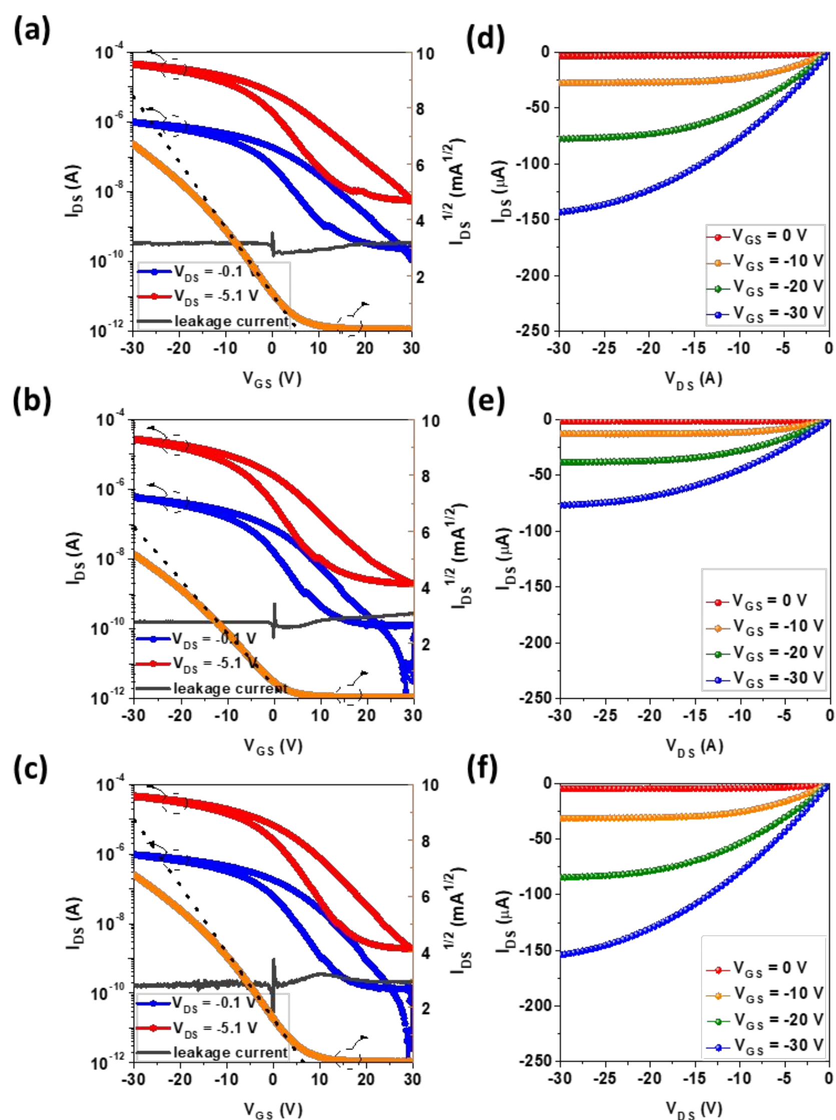

Figure S7. Transfer characteristics of the 1.9 at% La-loaded SnO TFTs annealed in 250 °C with different oxygen partial pressures of (a) 3.23 %, (b) 3.85 at%, and (c) 4.46 at%. The corresponding output characteristics of the 1.9 at% La-loaded SnO TFTs with oxygen partial pressures of (d) 3.23 %, (e) 3.85 %, and (f) 4.46 %.

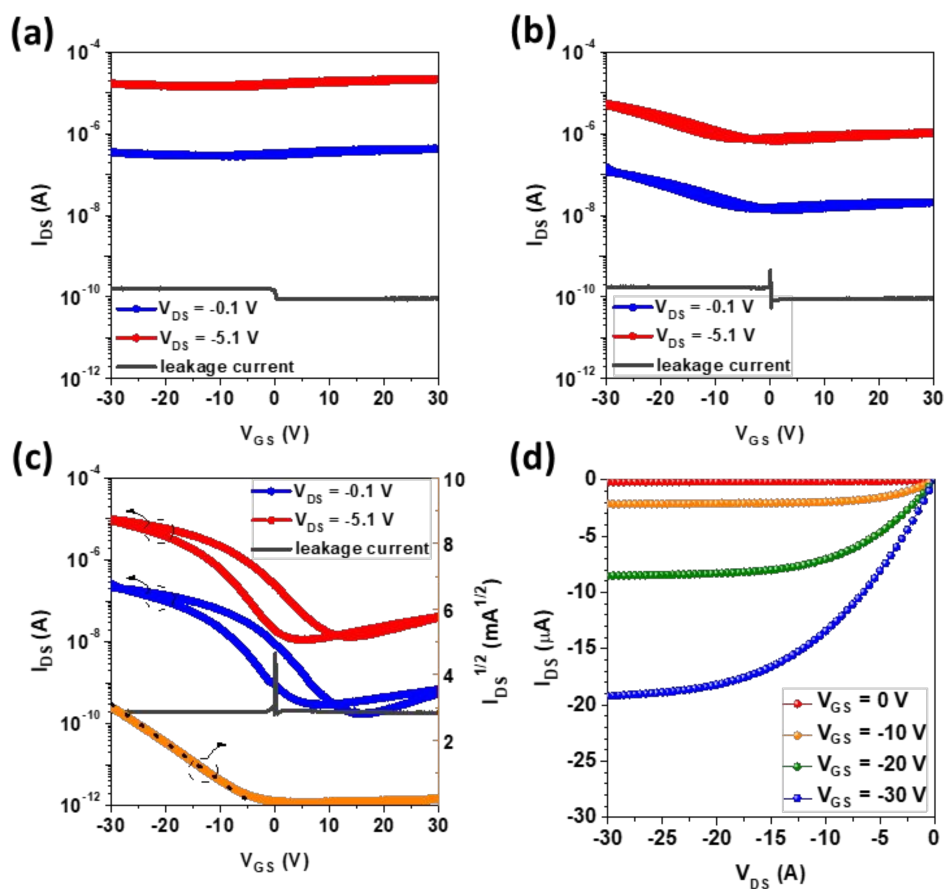

Figure S8. Transfer characteristics of the SnO TFTs annealed at 300 °C with different La loadings of (a) 0 at%, (b) 0.8 at%, and (c) 1.9 at%. (d) The output characteristics of the SnO TFTs with 1.9 at% La loadings

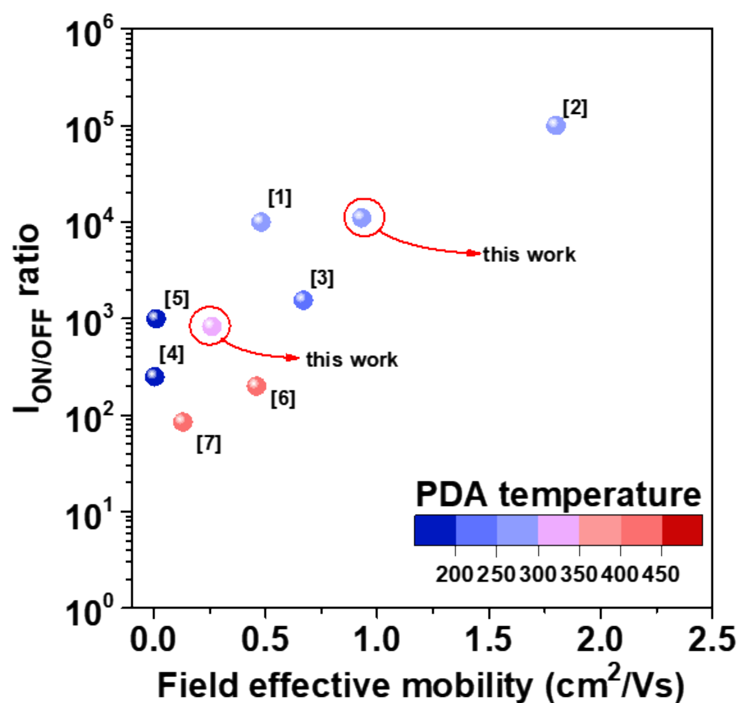

Figure S9. Comparisons of the trade-off between the saturation mobility and $I_{ON/OFF}$ ratio for the SnO TFTs fabricated at the various annealing temperatures. For fair comparison, the SnO TFTs with SiO_2 gate dielectric and non-passivated devices were collected. It can be shown that the performances of SnO TFTs reported in the literatures were deteriorated at the higher annealing temperature (> 300 °C) compared to those at the lower annealing temperature (< 300 °C). The introduction of La cation into the SnO channel allowed the resulting transistor to exhibit the higher mobility and $I_{ON/OFF}$ ratio due to the La-induced efficient suppression of Sn^{4+}, as shown in Figure 8b.

Table S1. Chemical compositions including Sn^0, Sn^{2+}, and Sn^{4+} of the La-free SnO films post-annealed at 250 ºC for 1 hr states, which was de-convoluted from XP spectra of Sn $3d_{5/2}$ and Sn $4d_{5/2}$.

XP Spectra	Sn^0 (%)	Sn^{2+} (%)	Sn^{4+} (%)
Sn $3d_{5/2}$	3.8	70.9	25.3
Sn $4d_{5/2}$	5.7	69.7	24.6

Table S2. Chemical compositions including Sn^0, Sn^{2+}, and Sn^{4+} of as deposited SnO films with different La loadings, which were de-convoluted from XP spectra of Sn 3d and O 1s.

La Loading in SnO film [at%]	Sn 3d			O 1s		
	Sn^{4+} (486.8 ± 0.1 eV)	Sn^{2+} (486.1 ± 0.1 eV)	Sn^0 (484.6 ± 0.1 eV)	O_{chem} (531.75 eV)	$O+Sn^{4+}$ (530.25 eV)	$O+Sn^{2+}$ (529.55 eV)
0	5.4	43.2	51.4	4.1	41.7	54.2
0.8	11.7	46.1	42.2	4.5	38.8	56.7
1.9	12.5	47.2	40.3	0.8	41.1	58.1
3.1	13.7	53.0	33.3	4.4	34.0	61.6

Table S3. Chemical compositions including Sn^0, Sn^{2+}, and Sn^{4+} of SnO films annealed at 250 °C with different La loadings, which were de-convoluted from XP spectra of Sn 3d and O 1s.

La Loading in SnO film [at%]	Sn 3d			O 1s		
	Sn^{4+} (486.4 ± 0.3 eV)	Sn^{2+} (485.7 ± 0.3 eV)	Sn^0 (484.2 ± 0.3 eV)	O_{chem} (531.75 eV)	$O+Sn^{4+}$ (530.25 eV)	$O+Sn^{2+}$ (529.55 eV)
0	25.3	70.9	3.8	3.7	27.4	68.9
0.8	23.4	74.2	2.4	3.8	23.1	73.1
1.9	16.9	79.7	3.4	3.9	17.4	78.7
3.1	19.2	65.4	15.4	3.1	33.7	63.2

Table S4. Hall effect measurements for the SnO films with different La loadings of 0 and 1.9 at%, which were annealed in forming gas atmosphere in 250 °C.

La Loading in SnO film [at%]	majority carrier type	μ_{Hall} (cm^2/Vs)	N_{Hall} (cm^{-3})
0	electron	1.2 ± 0.9	$4.0\ (\pm 2.7) \times 10^{18}$
1.9	electron	3.5 ± 3.7	$1.7\ (\pm 0.9) \times 10^{19}$

Table S5. Chemical compositions including Sn^0, Sn^{2+}, and Sn^{4+} of SnO films annealed at 250 °C with different oxygen pressure on 1.9 at% La loadings, which were de-convoluted from XP spectra of Sn 3d and O 1s.

Oxygen partial pressure [%]	Sn 3d			O 1s		
	Sn^{4+} (486.4 ± 0.3 eV)	Sn^{2+} (485.7 ± 0.3 eV)	Sn^{0} (484.2 ± 0.3 eV)	O_{chem} (531.75 ± 0.2 eV)	$O+Sn^{4+}$ (530.25 ± 0.2 eV)	$O+Sn^{2+}$ (529.55 ± 0.2 eV)
3.23	15.3	81.7	3.0	3.9	17.4	78.7
3.85	3.8	90.6	5.6	2.7	4.8	92.5
4.46	5.9	92.2	1.9	1.1	5.3	93.6
5.06	5.7	90.5	3.8	1.1	9.2	89.7

Reference

(1) Nomura, K.; Kamiya, T.; Hosono, H. Ambipolar Oxide Thin-Film Transistor. *Adv. Mater.* **2011**, *23*, 3431–3434.

(2) Han, Y.; Choi, Y.; Jeong, C.; Lee, D.; Song, S.-H.; Kwon, H.-I. Environment-Dependent Bias Stress Stability of *p*-Type SnO Thin-Film Transistors. *IEEE Electron Device Lett.* **2015**, *36*, 466–468.

(3) Luo, H.; Liang, L. Y.; Liu, Q.; Cao, H. T. Magnetron-Sputtered SnO Thin Films for *p*-Type and Ambipolar TFT Applications. *ECS J. Solid State Sci. Technol.* **2014**, *3*, Q3091–Q3094.

(4) Dhananjay; Chu, C.-W.; Ou, C.-W.; Wu, M.-C.; Ho, Z.-Y.; Ho, K.-C.; Lee, S.-W. Complementary Inverter Circuits Based on *p*-SnO_2 and *n*-In_2O_3 Thin Film Transistors. *Appl. Phys. Lett.* **2008**, *92*, 232103.

(5) Ou, C.-W.; Dhananjay; Ho, Z. Y.; Chuang, Y.-C.; Cheng, S.-S.; Wu, M.-C.; Ho, K.-C.; Chu, C.-W. Anomalous *p*-Channel Amorphous Oxide Transistors Based on Tin Oxide and Their Complementary Circuits. *Appl. Phys. Lett.* **2008**, *92*, 122113.

(6) Liang, L. Y.; Liu, Z. M.; Cao, H. T.; Yu, Z.; Shi, Y. Y.; Chen, A. H.; Zhang, H. Z.; Fang, Y. Q.; Sun, X. L. Phase and Optical Characterizations of Annealed SnO Thin Films and Their *p*-Type TFT Application. *J. Electrochem. Soc.* **2010**, *157*, H598–H602.

(7) Okamura, K.; Nasr, B.; Brand, R. A.; Hahn, H. Solution-Processed Oxide Semiconductor SnO in *p*-Channel Thin-Film Transistors. *J. Mater. Chem.* **2012**, *22*, 4607–4610.