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Supporting Information

Reversible Redox Control of Optoelectronic Properties of Hexagonal Tungsten Oxide Epitaxial Films Grown on YSZ Solid Electrolyte

Gwoon Kim*, Hai jun Cho, and Hiromichi Ohta*

¹Graduate School of Information Science and Technology, Hokkaido University, N14W9, Kita, Sapporo 060-0814, Japan

²Research Institute for Electronic Science, Hokkaido University, N20W10, Kita, Sapporo 001-0020, Japan

*To whom correspondence should be addressed: G.K. (woom93@gmail.com), H.O. (hiromichi.ohta@es.hokudai.ac.jp)

Table S1. Transmission at 1.5 μm (*Trans.*) and the activation energy (E_a) of the electrochemically reduced/oxidized h-WO_x films. The E_a was extracted by assuming the Arrhenius-type thermal activation of σ as $\sigma = \sigma_0 \exp(-E_a/k_B T)$, where σ_0 , k_B and T are pre-exponential factor, Boltzmann constant, and absolute temperature, respectively.

| Sample | x in WO_x | <i>Trans.</i> at 1.5 μm (%) | E_a (meV) |
|---------------|---|--|-------------------------------|
| A | 2.987 | 49.8 | 212.3 |
| B | 2.986 | 52.0 | 203.7 |
| C | 2.981 | 46.7 | 108.5 |
| D | 2.960 | 44.2 | 74.0 |
| E | 2.954 | 43.4 | 68.1 |
| F | 2.935 | 41.8 | 71.7 |
| G | 2.933 | 38.3 | 35.8 |
| H | 2.931 | 35.4 | 33.3 |
| Oxi 1 | 2.993 | 69.3 | ND |
| Oxi 2 | 2.988 | 61.9 | ND |

Table S2. *Trans.* and E_a of the PLD-grown h-WO_x films.

| Sample | x in WO_x | <i>Trans.</i> at 1.5 μm (%) | E_a (meV) |
|---------------|---|--|-------------------------------|
| 6 Pa | 2.993 | 54.9 | 223.5 |
| 5 Pa | 2.984 | 51.6 | 99.5 |
| 4 Pa | 2.980 | 45.2 | 74.4 |
| 3 Pa | 2.957 | 35.5 | 41.6 |
| 2 Pa | 2.938 | 34.4 | 18.1 |

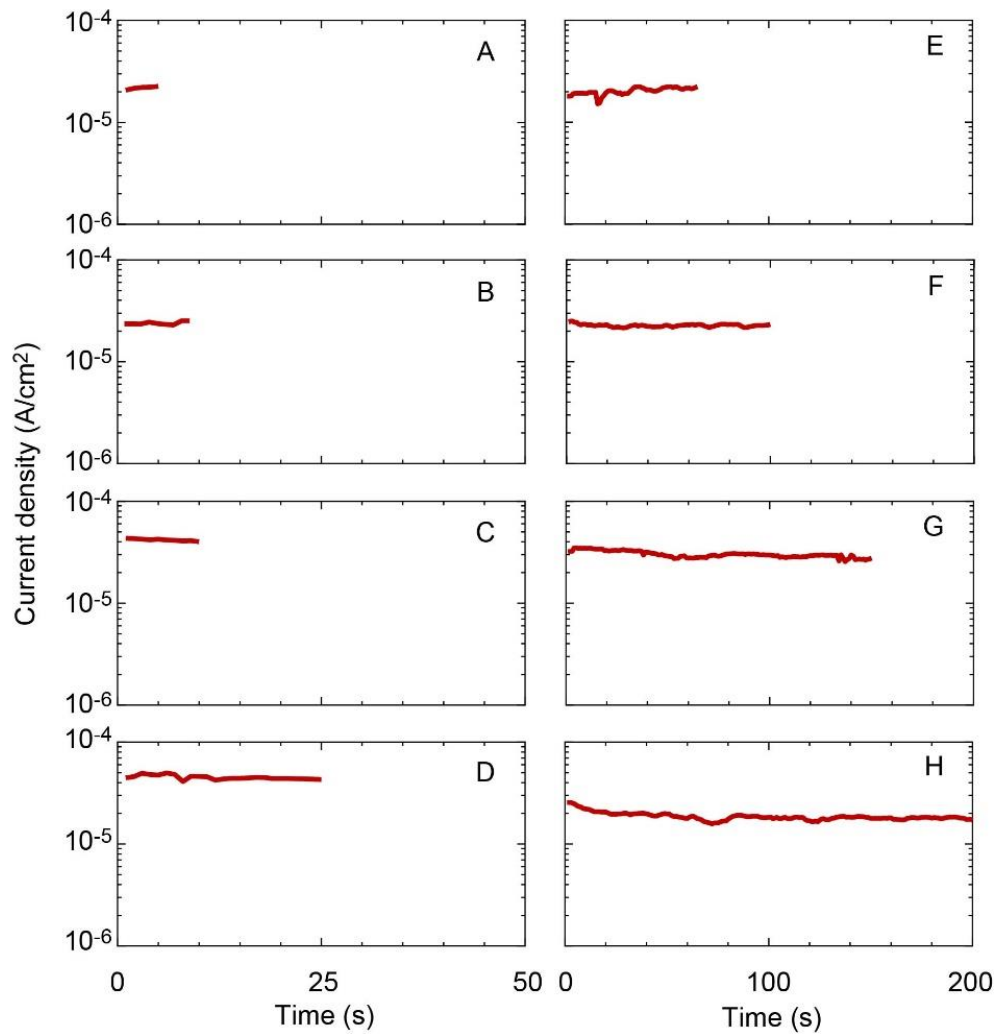


Figure S1. Time dependence of the current density during the electrochemical reduction treatment by applying +3 V to YSZ substrate.

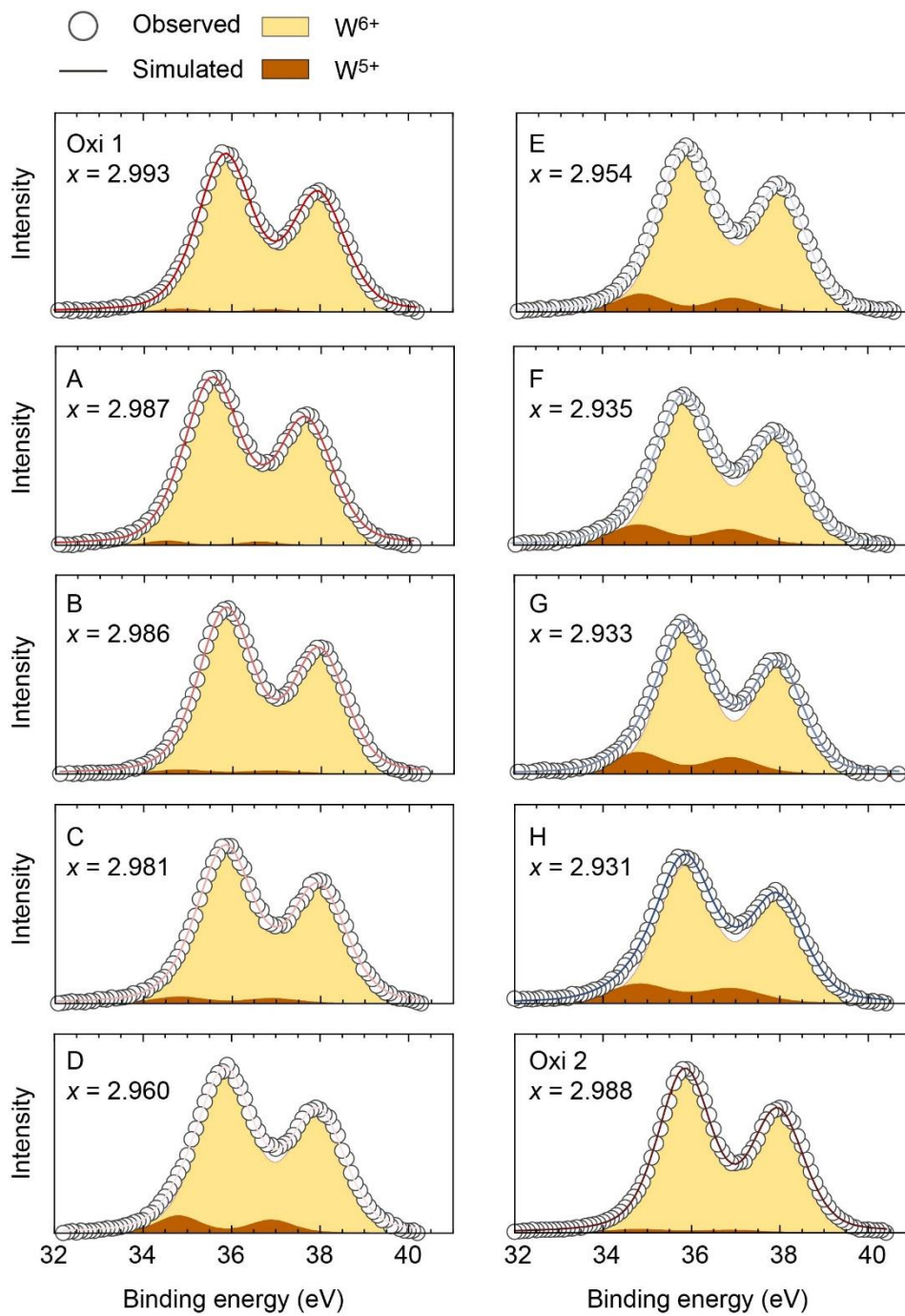


Figure S2. XPS spectra around W 4f peaks of the electrochemically redox treated h-WO_x epitaxial films.

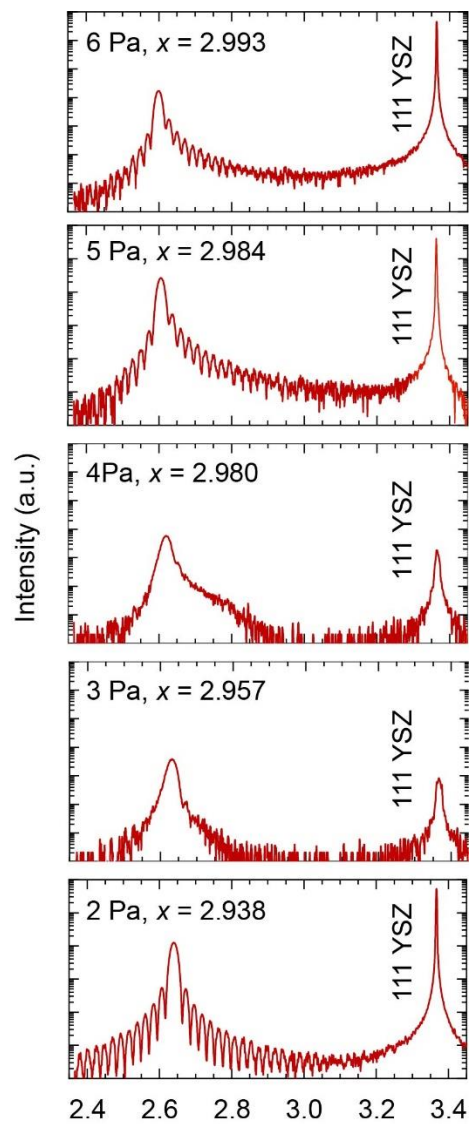


Figure S3. Change in the out-of-plane XRD patterns of PLD-grown h-WO_x epitaxial films under various oxygen pressure from 2 to 6 Pa during the film growth.

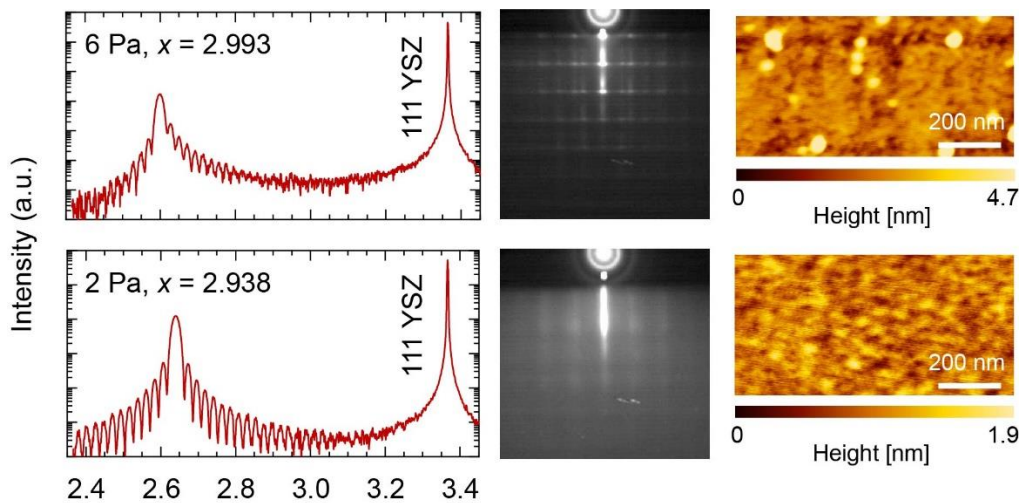


Figure S4. Comparison of the crystallographic feature of the PLD-grown $h\text{-WO}_x$ epitaxial films under oxygen pressure of 6 Pa and 2 Pa. (from left to right: out-of-plane XRD patterns, RHEED patterns, and topographic AFM images)

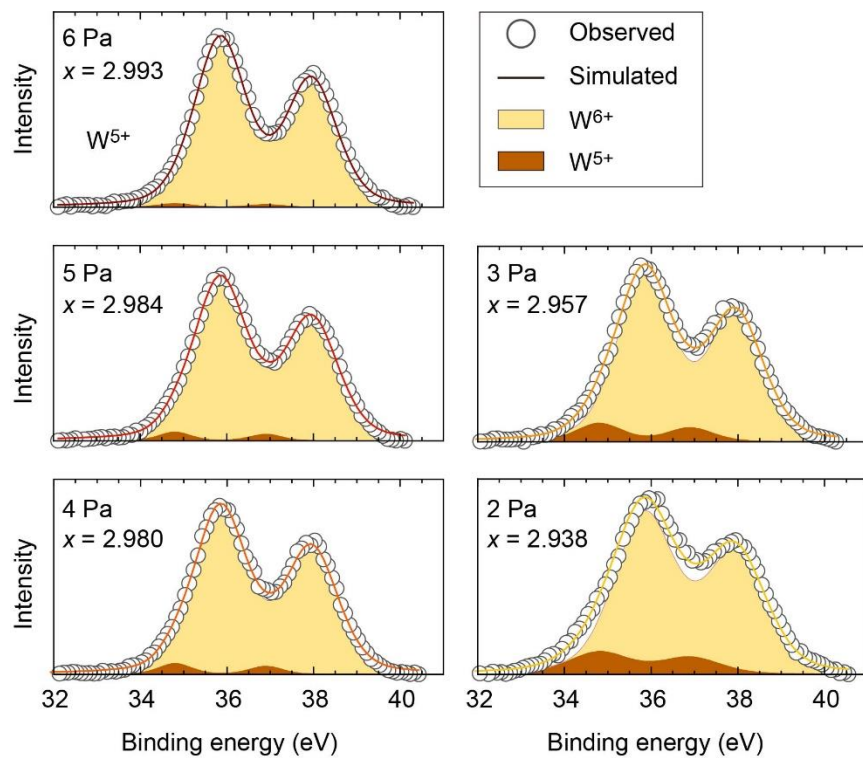


Figure S5. XPS spectra around W 4f peaks of PLD-grown $h\text{-WO}_x$ epitaxial films under various oxygen pressure from 2 to 6 Pa during the film growth.

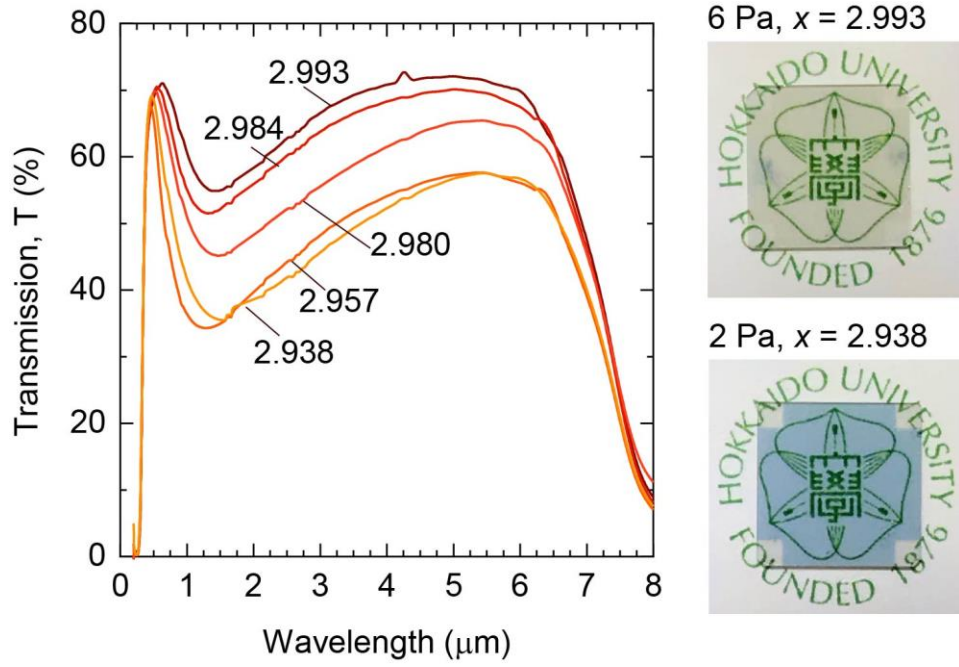


Figure S6. Optical transmission and photographs of PLD-grown $h\text{-WO}_x$ epitaxial films under various oxygen pressure from 2 to 6 Pa during the film growth.

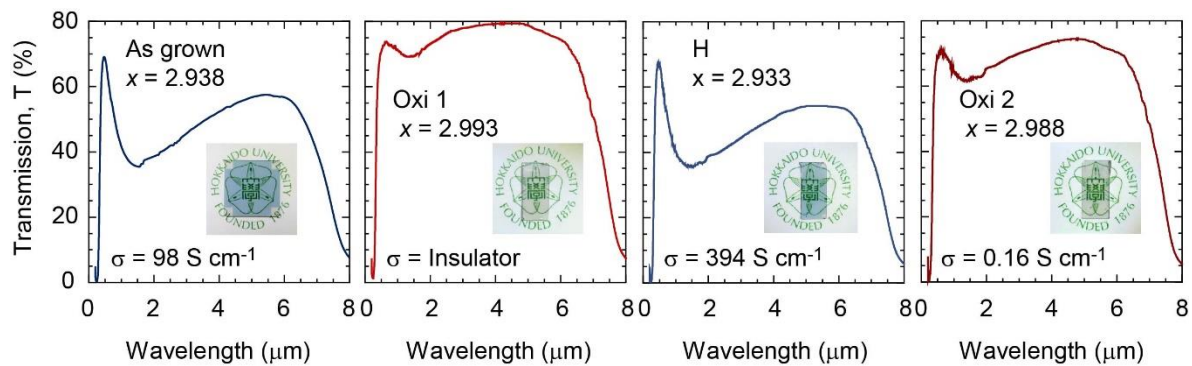


Figure S7. Changes in the optical transmission spectra of the h-WO_x epitaxial film after repeated electrochemical redox treatment. (From left to right: as-grown, 1st oxidized, 1st reduced, and 2nd oxidized)

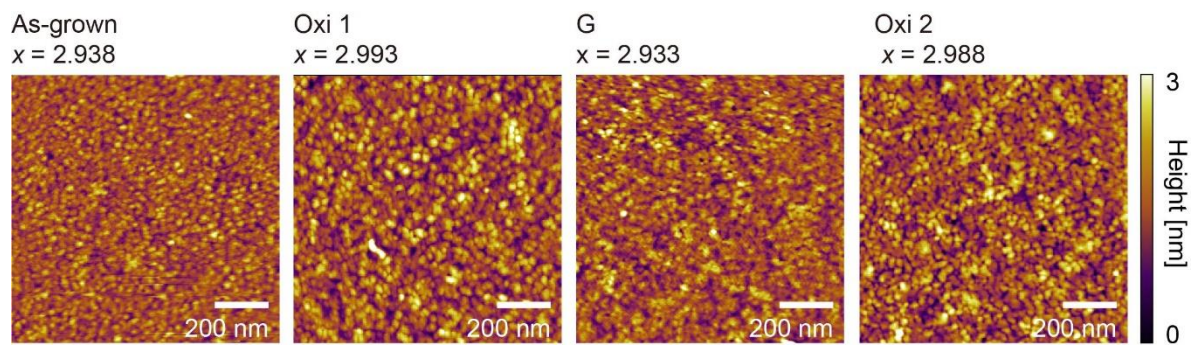


Figure S8. Surface morphology of the h-WO_x epitaxial film after repeated electrochemical redox treatment. (From left to right: as-grown, 1st oxidized, 1st reduced, and 2nd oxidized)