

Supplementary Information

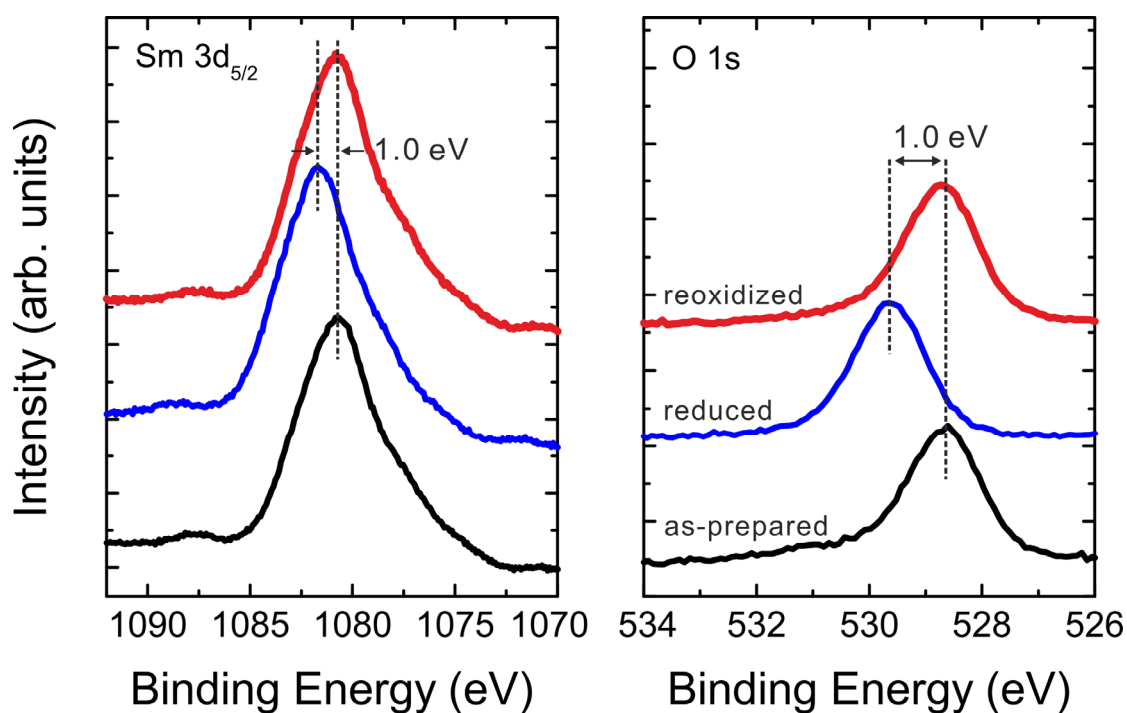


Figure S1. XPS spectra of Sm 3d_{5/2} and O 1s regions collected from the 2.8 ML as-prepared, reduced, and reoxidized SmO_x thin films.

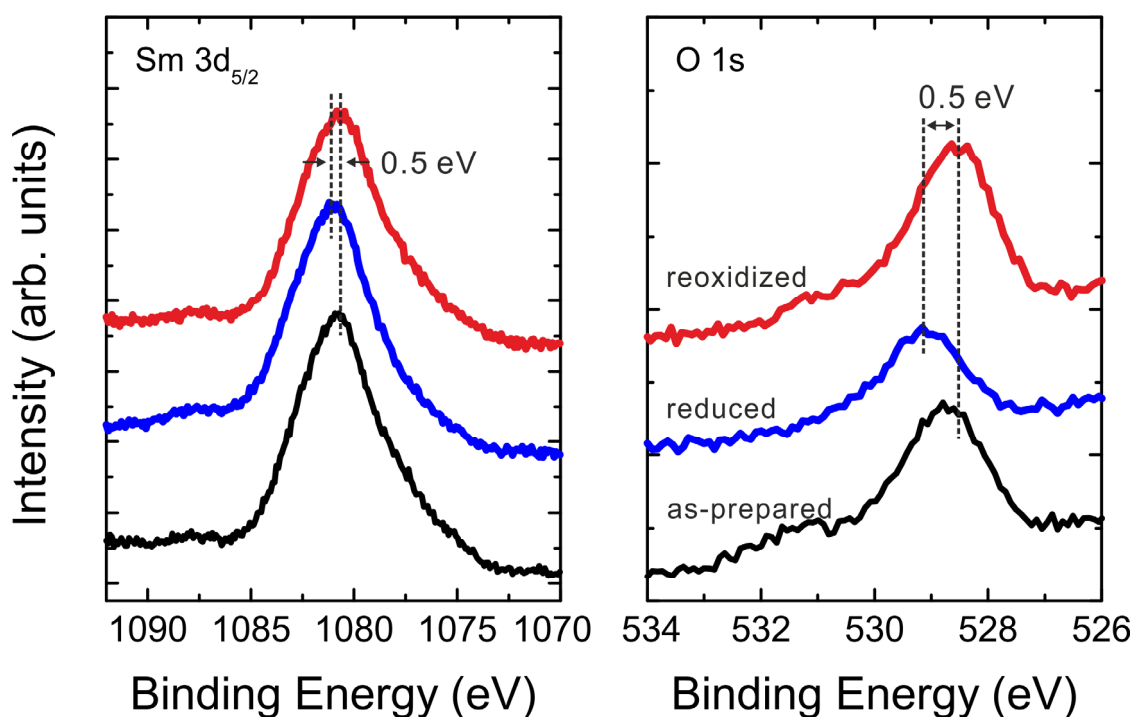
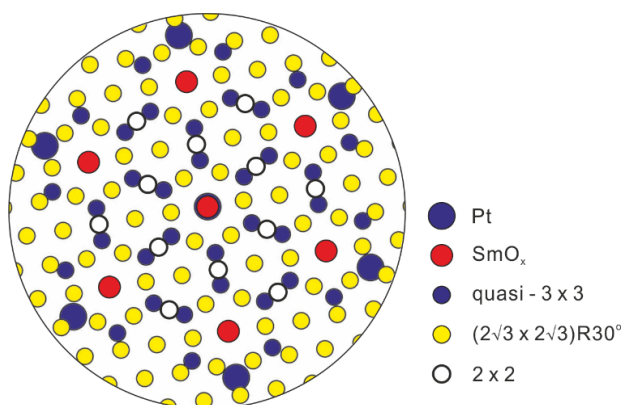


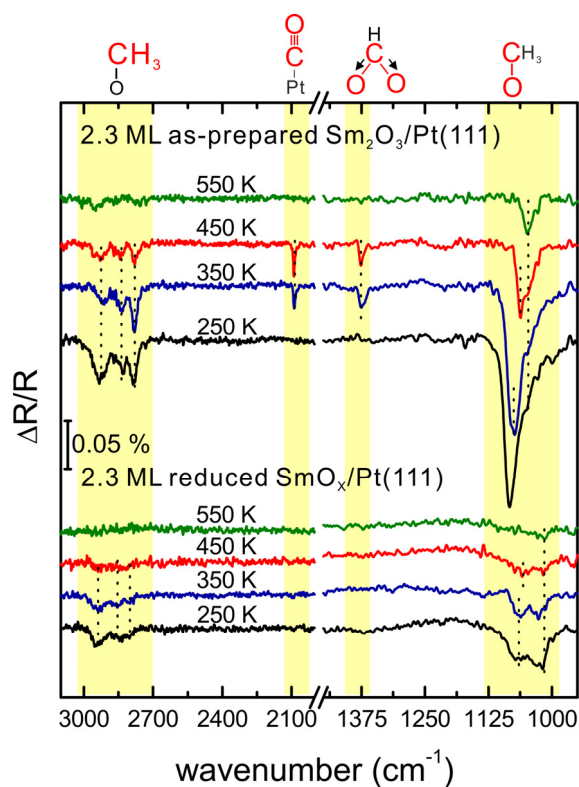
Figure S2. XPS spectra of Sm 3d_{5/2} and O 1s regions collected from the 0.9 ML as-prepared, reduced, and reoxidized SmO_x thin films.

Schematic representation of LEED patterns

**Figure S3.** Summary of LEED patterns observed.**Table S1.** Summary of LEED patterns observed for the differently prepared SmO_x deposits.

MeOH TPD		LEED Patterns of 0.9 ML SmO_x on Pt(111)		
Treatment	as-prepared Sm_2O_3	reduced SmO_x	reoxidized Sm_2O_3	
w/o	quasi- 3×3	satellites	quasi- $3 \times 3 + 2$ add. spots	
with	slightly rotated	rotated	quasi- $3 \times 3 + 2$ add. spots + $(2\sqrt{3} \times 2\sqrt{3})\text{R}30^\circ$	

MeOH TPD		LEED Patterns of 2.8 ML SmO_x on Pt(111)		
Treatment	as-prepared Sm_2O_3	reduced SmO_x	reoxidized Sm_2O_3	
w/o	quasi- 3×3	quasi- $3 \times 3 + (2\sqrt{3} \times 2\sqrt{3})\text{R}30^\circ$	quasi- $3 \times 3 + 2 \times 2$	
with	$\text{SmO}_x(1 \times 1)$	quasi- $3 \times 3 + (2\sqrt{3} \times 2\sqrt{3})\text{R}30^\circ$	quasi- $3 \times 3 + (2\sqrt{3} \times 2\sqrt{3})\text{R}30^\circ$	

**Figure S4.** Temperature-dependent IRRAS spectra collected from the 2.3 ML as-prepared and reduced SmO_x thin films.