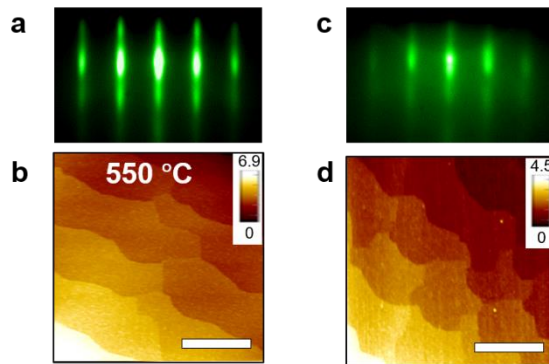


Carbonate formation lowers the electrocatalytic activity of perovskite oxides for water electrolysis

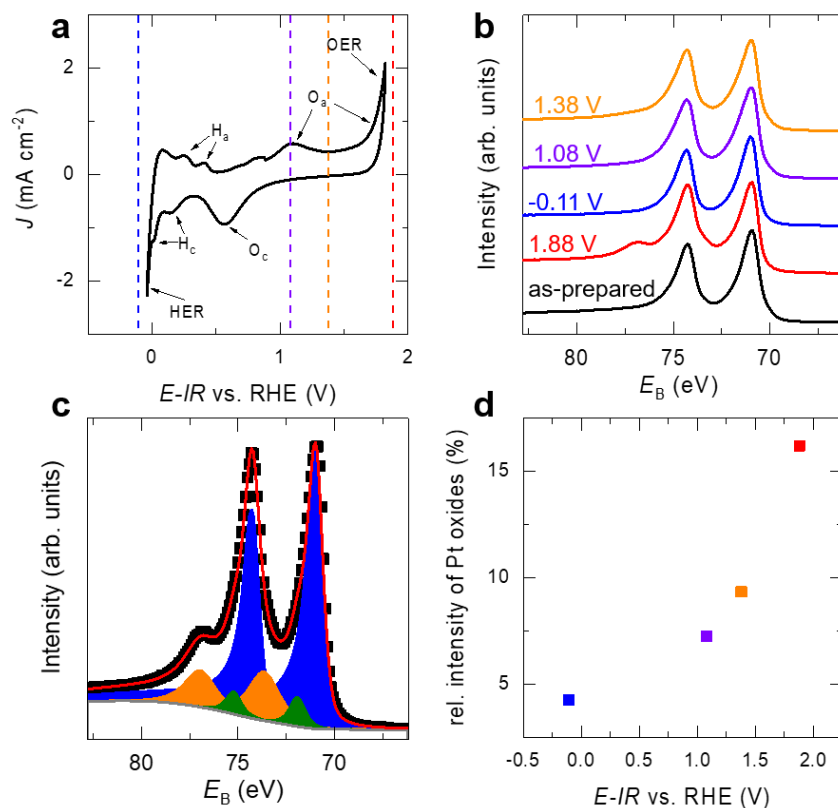
SUPPLEMENTARY INFORMATION

Christoph Baeumer^{1,2,3,4*}, *Allen Yu-Lun Liang*^{2,5}, *Urška Trstenjak*^{6‡}, *Qiyang Lu*^{1,2,7}, *Rainer Waser*^{3,6}, *J. Tyler Mefford*^{1,2}, *Felix Gunkel*⁶, *Slavomír Nemšák*^{6,7*}, *William C. Chueh*^{1,2}

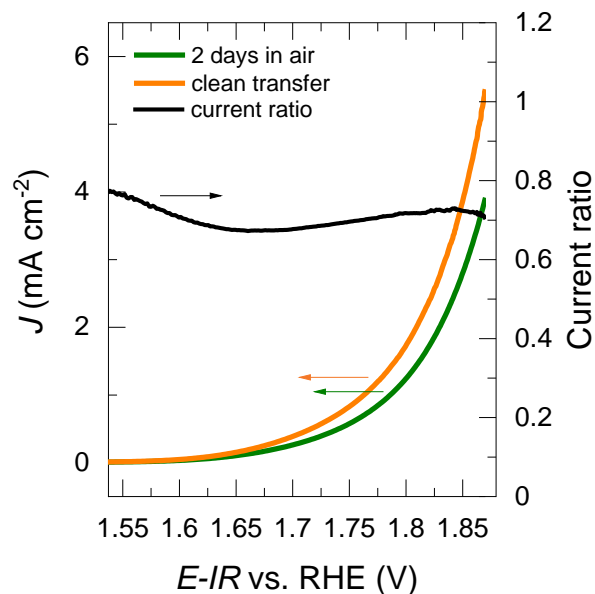
- 1 Department of Materials Science and Engineering, Stanford University, Stanford, CA 94305, USA
 - 2 Stanford Institute for Materials and Energy Science, SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA
 - 3 Institute of Electronic Materials (IWE2) and JARA-FIT, RWTH Aachen University, 52074 Aachen, Germany
 - 4 MESA+ Institute for Nanotechnology, University of Twente, Faculty of Science and Technology, 7500 AE Enschede, Netherlands
 - 5 Department of Chemistry, Stanford University, Stanford, CA 94305, USA
 - 6 Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA
 - 7 Peter Gruenberg Institute and JARA-FIT, Forschungszentrum Juelich GmbH, 52425 Juelich, Germany
- ‡ Present address: Advanced Materials Department, Jožef Stefan Institute, SI-1000 Ljubljana, Slovenia



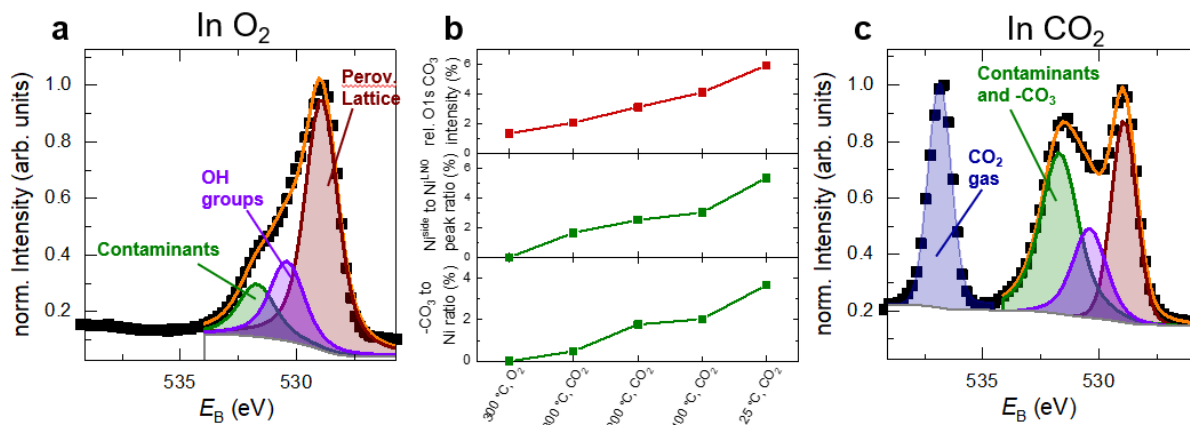
Supplementary Figure 1: (a) RHEED pattern after growth. (b) Atomic force microscopy after UHV-transfer to the AFM chamber. (c) RHEED pattern after electrochemical characterization. The intensity of the electron beam was identical and all settings as similar to panel a as possible. The loss in intensity of the diffraction spots compared to the background intensity indicates a slight loss of surface crystallinity. (d) Atomic force microscopy after electrochemical characterization.



Supplementary Figure 2: (a) Cyclic voltammetry of a 50 nm Pt layer at 500 mV/s. The redox features are labeled according to Bard and Faulkner.¹⁹ H_c/H_a: formation/oxidation of adsorbed hydrogen. O_a/O_c: formation/reduction of adsorbed oxygen or a platinum oxide layer. (b) Pt 4f XPS after deposition and after potential holds at different potentials as indicated in panel a. First, the film was oxidized at 1.88 V vs. RHE, then it was reduced at -0.11 V vs. RHE, followed by reoxidation at 1.08 and 1.38 V vs. RHE, respectively. (c) representative XPS fitting of the Pt 4f level. (d) Relative intensity of the Pt oxide peaks as a function of applied potential.



Supplementary Figure 4: (a) Cyclic voltammety of the same films as in Figure 2 of the main text and current normalization: The current ratio was obtained by dividing the data from the air-exposed sample by the clean-transfer data. The ratio is fairly constant at ~ 70 to 80 %.



Supplementary Figure 3: (a) O1s spectrum of a LaNiO_3 film in O_2 at 300 °C. The surface is clean except for a small contamination peak often observed for perovskite oxides (doi/10.1021/acs.chemmater.9b05151) (b) Top panel: Increase of the peak corresponding to the O $1s^{\text{CO}_3}$ and contaminations during cooling down in CO_2 . Based on their binding energy, these peaks could not be separated unambiguously. Middle and bottom panel: Intensity ratio of the C $1s^{\text{CO}_3}$ peak and the total Ni $3p$ intensity (same data as shown in Fig. 5 of the main text). (c) O1s spectrum of the same film after cooling down to room temperature in CO_2 .