and Ni2. The Ni1–O1–Ni2 bond angle is about 145° and the length of the Ni2–O2 bond is approximately 2.5 Å, similar to that of bulk SNO. The proton first rotates about O1 while being bound to it, so that it enters the sub-surface layer of the SNO slab and reduces the O2–H distance to about 2.6 Å (image I_2 in Fig. 3c). This rotation of the O1–H bond distorts the surface layer considerably, which manifests itself in an increased Ni2–O2 distance of about 3.43 Å and a substantial change of the Ni1–O1–Ni2 angle (about 153°). This surface distortion process is associated with a barrier of about 0.9 eV. Further rotation of the O1–H bond brings the proton close to O2 (O2–H distance of approximately 1.5 Å; image I_3 in Fig. 3c) while increasing the inter-octahedral angle Ni1–O1–Ni2 to about 175°, and leads to the concurrent healing of the Ni2–O2 bond (image I_4 in Fig. 3c), which thus preserves the SNO framework upon intake of protons. Once the proton is intercalated into the SNO lattice, intra-octahedral proton hopping occurs even at room temperature, as seen in the *ab initio* simulations (Supplementary Video 3), indicating facile proton diffusion within SNO. The calculated energy barrier associated with the migration of protons within the SNO lattice is about 0.27 eV, which is low compared to those of other proton-conducting oxides[28] (0.4–0.6 eV). Volume expansion is observed in simulations after proton intercalation (Extended Data Fig. 10a–g and Supplementary Information section 7), which is in agreement with reflectometry measurements.

Density functional theory calculations (Fig. 3d) show that as each hydrogen atom is added to the supercell, its electron is transferred to a previously unoccupied Ni–O orbital of pristine SNO. The transfer of the electron can also be seen in the difference between the charge density of the combined HSNO system and that of the sum of SNO and H, which shows charge depletion around the H and a corresponding charge accumulation around the adjacent Ni and O (Extended Data Fig. 10h, i). Owing to electron–electron correlations, this state is shifted down into the valence band, while the remaining unoccupied states of that Ni are shifted up in energy. The bandgap remains almost unchanged until an electron has been added to each Ni atom. Both of these observations are consistent with the experimentally observed changes in X-ray absorption spectra and the increase in the electrical resistance of SNO that enables effective sensing.

Online Content Methods, along with any additional Extended Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.

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Supplementary Information is available in the online version of the paper.

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Author Contributions Z.Z., D.S. and S.R. conceived the study. Z.Z. and D.S. fabricated the SNO thin-film devices and performed the electrical, stability, sensing and electrochemical experiments. B.N., M.C. and S.K.R.S.S. performed the AIMD simulations and the nudged elastic band calculations to compute activation barriers. M.K. and K.M.R. performed first-principles electronic structure calculations. D.S. and J.A.D. performed neutron reflectivity measurements. Y.S., D.S. and H.Z. performed X-ray diffraction and X-ray reflectivity measurements. J.W.F., J.L., R.S., F.H. and R.C. performed X-ray absorption measurements. C.W. and N.Y. performed optical measurements and analysis. J.Z. and S.S.N. performed cross-sectional conducting AFM studies. K.R. and Z.Z. performed in-plane conducting AFM studies. Z.Z., D.S., B.N., S.K.R.S.S. and S.R. wrote the manuscript. All authors discussed the results and commented on the manuscript.

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