

crystallographic direction [110]. Periodic boundary conditions were employed along all directions with a vacuum of about 10 Å along the surface normal. For the SNO/water simulations, this vacuum was filled with 18 water molecules to simulate the experimental water density (1 g cm^{-3}). The exchange correlation was described by the Perdew–Burke–Ernzerhof functional³³, with the pseudopotentials Sm_3 (valence $5s^2 5p^2 6s^2 4f^4$), Ni_pv (valence $3p^6 4s^2 3d^8$) and O (valence $2s^2 2p^4$) supplied by VASP. The plane-wave energy cut-off was set at 520 eV. The Brillouin zone was sampled at the Γ -point only. Using AIMD simulations in the isobaric-isothermal ensemble, we first thermalized the SNO (110) computational supercell at various temperatures ranging from 300 K to 500 K and zero external pressure for 10 ps using a time step of 0.5 fs. During these simulations, the cell volume, cell shape and atomic positions were allowed to vary via the Parrinello–Rahman scheme³⁴. The temperature conditions were maintained by using a Langevin thermostat. Next, we inserted the water molecules in the vacuum (at a given temperature). The subsequent AIMD simulations were performed in the canonical ensemble (constant volume and temperature). Constant temperature conditions were maintained via a Nosé–Hoover thermostat³⁵, as implemented in VASP. To identify the activation barriers and minimum energy paths for H intercalation into a SNO (110) slab, we employed the climbing image nudged elastic band method within the GGA + U formalism³⁶, where U is the on-site Coulomb parameter. The diffusion coefficient (D) of protons in bulk SNO at 300 K was estimated using the Einstein relationship $D = [r(t) - r(0)]^2 / (6t)$, where $[r(t) - r(0)]^2$ is the mean-square displacement of a proton at time t with respect to the time origin ($t = 0$); the value of D was averaged over various time domains (each of duration 0.5 ps) over the AIMD trajectory.

First-principles electronic structure calculations of HSNO. First-principles calculations were carried out within the DFT + U approximation with the VASP code^{31,32} using the projector augmented plane-wave method of DFT³⁷ and the pseudopotentials Sm_3 (valence $5s^2 5p^2 6s^2 4f^4$), Ni_pv (valence $3p^6 4s^2 3d^8$), O (valence $2s^2 2p^4$) and H (valence $1s^1$). To treat the exchange and correlation, the Perdew–Burke–Ernzerhof functional was used within the GGA³³ and the rotationally invariant form of DFT + U of ref. 34 with $U = 4.6 \text{ eV}$ and $J = 0.6 \text{ eV}$, where J is the on-site exchange parameter. For the structural determination of pristine SNO, we started with the Materials Project structure³⁸, added a small monoclinic distortion ($\beta \approx 90.75^\circ$) and allowed the cell and ionic positions to relax until the forces on each ion were lower than $0.005 \text{ eV \AA}^{-1}$. All calculations were carried out with the tetrahedral method with Blöchl corrections³⁹, a $6 \times 6 \times 4$ Monkhorst–Pack k -point mesh for the $\sqrt{2} \times \sqrt{2} \times 2$ supercell, and a plane-wave energy cut-off of 500 eV. To determine the structure of HSNO, we began with $\text{H}_{0.25}\text{SmNiO}_3$, adding one hydrogen atom at various locations to the pristine SNO $\sqrt{2} \times \sqrt{2} \times 2$ supercell with G-type magnetic ordering, and allowed the internal coordinates to relax with the same tolerance as described above. The structure with the lowest energy

was chosen as the $\text{H}_{0.25}\text{SmNiO}_3$ structure. Taking the symmetry-equivalent site of the relaxed hydrogen position, we constructed structures for $\text{H}_{0.5-1}\text{SmNiO}_3$, again allowing the internal coordinates to relax (Supplementary Information section 7). We compared the results while keeping the total volume of the cell fixed and relaxing the volume and the [110] direction only, where the qualitative features of the electronic structure were not affected.

Data availability. The data that support the findings of this study are available from the corresponding author upon reasonable request.

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