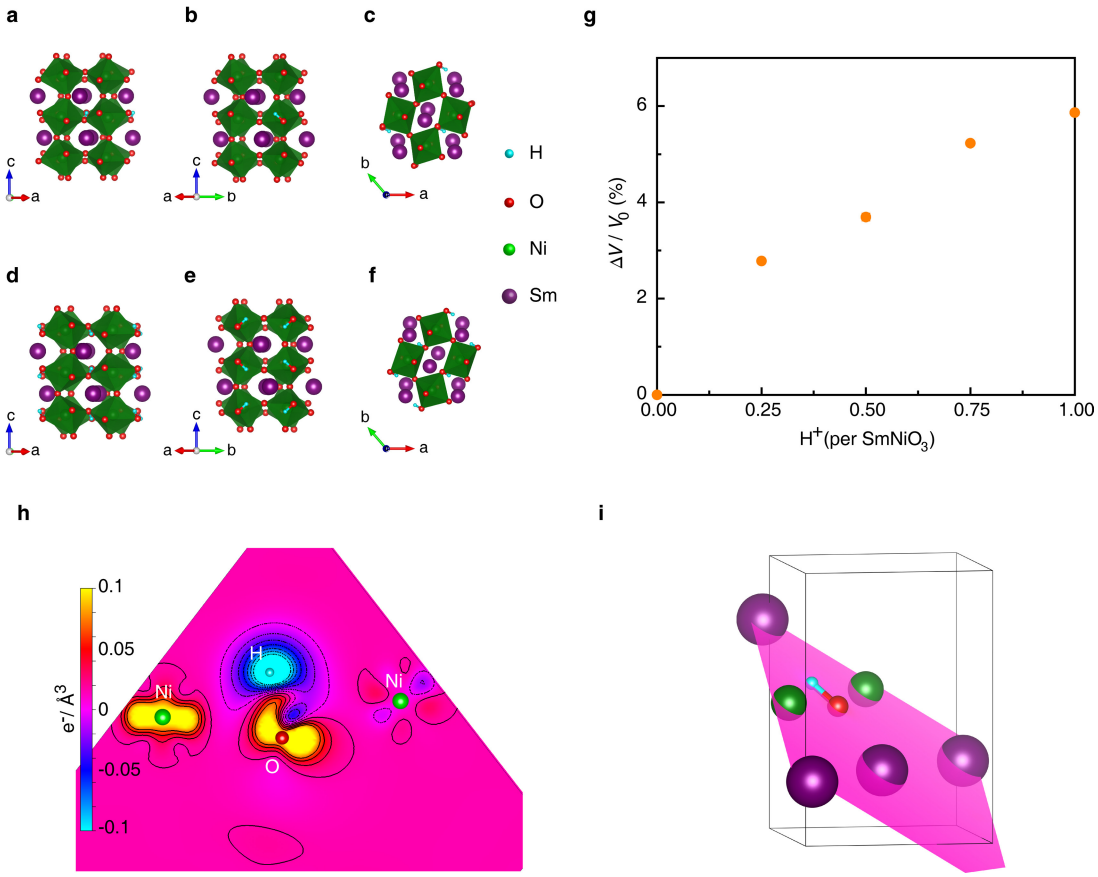

Extended Data Figure 10 | First-principles calculations of the structure and electron localization of HSNO. a–f, The structure of SNO with 1/4 H/SNO (**a–c**) and 1 H/SNO (**d–f**), displayed along the three crystallographic axes of the primitive perovskite structure. The crystallographic axes of the supercell were used in the calculations, where the [110] direction was allowed to relax. In all panels, 12 NiO_6 octahedra encompassing the Ni atoms (green) are displayed, with O in red, Sm in purple and H in cyan. The calculations use a $\sqrt{2} \times \sqrt{2} \times 2$ supercell (that is, with four Ni atoms). **g**, Change in the volume of SNO at various protonation levels (denoted as protons per SNO formula unit) obtained from DFT calculations. The calculated volume expansion for 1 H/SNO is about 5.9%, which is close to the value obtained from neutron reflectometry measurements and X-ray diffraction. **h**, The difference in the electron density between the relaxed HSNO ($SmNiO_3H$) and the initial state (SNO + H), which clearly shows a depletion (cyan) of charge around the hydrogen (cyan) and an accumulation (yellow) of charge around the closest nickel (green) and oxygen (red), which are part of the octahedron that expands upon hydrogen incorporation into the lattice. In this calculation, the *c* axis was allowed to relax while the other two (in-plane lattice constants) were fixed. For clarity, only the spin-down charge density is plotted because the electron incorporation results in a negative total magnetic moment (see the projected density of states of 1/4 H/SNO in Fig. 3d). **i**, The $(\bar{1}11)$ plane of the contour plot situated within the supercell.