Supplementary information

Enhancing electrochemical methane coupling in solid oxide cells by tuning oxygen species in the catalyst

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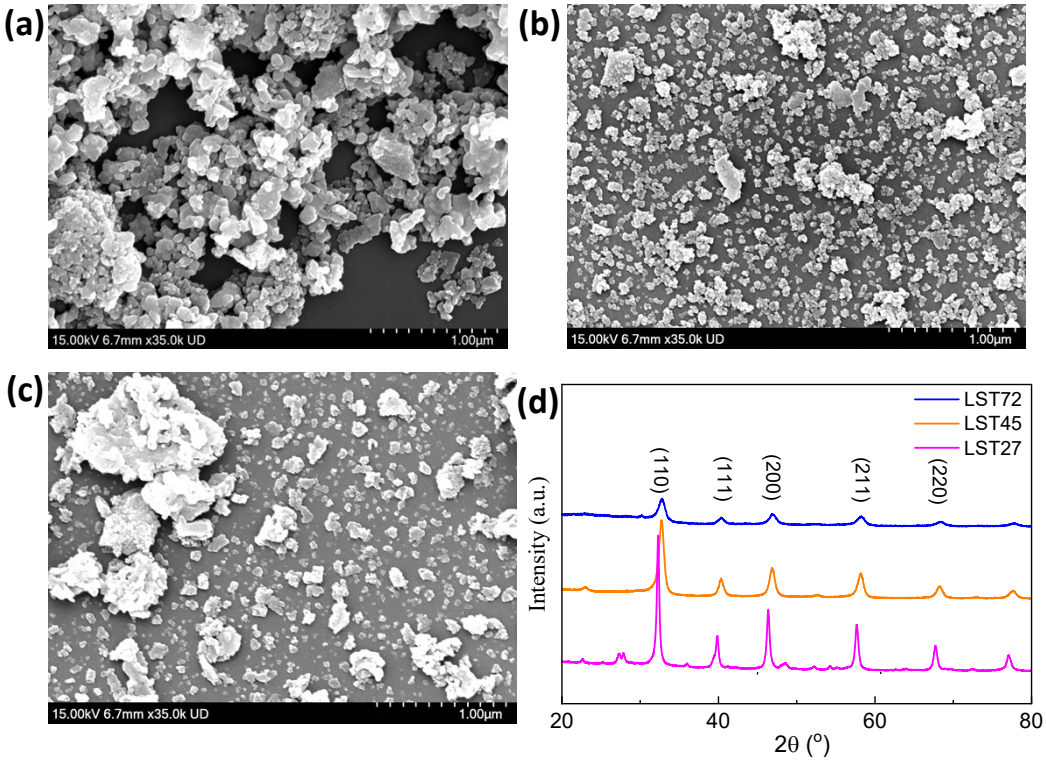

Fig. S1 Morphology of LST powder (a) LST27; (b) LST45; (c) LST72; (d) XRD patters for LST.

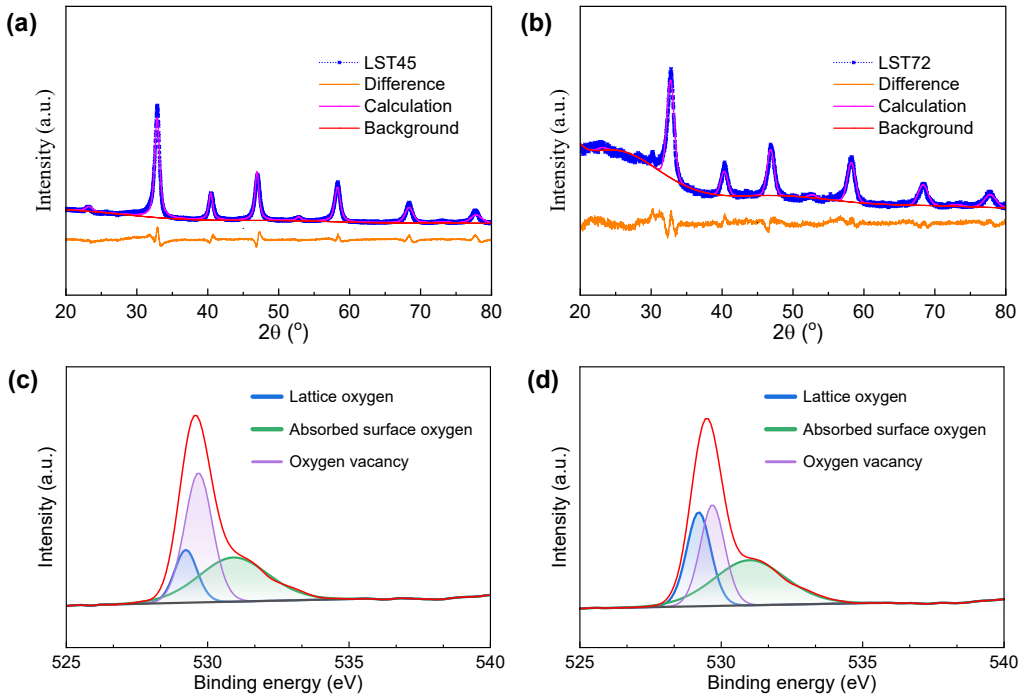

Fig. S2 X-ray diffraction Rietveld refinements of LST powder (a) LST45; (b) LST72; XPS for LST (c) LST45; (d) LST72.

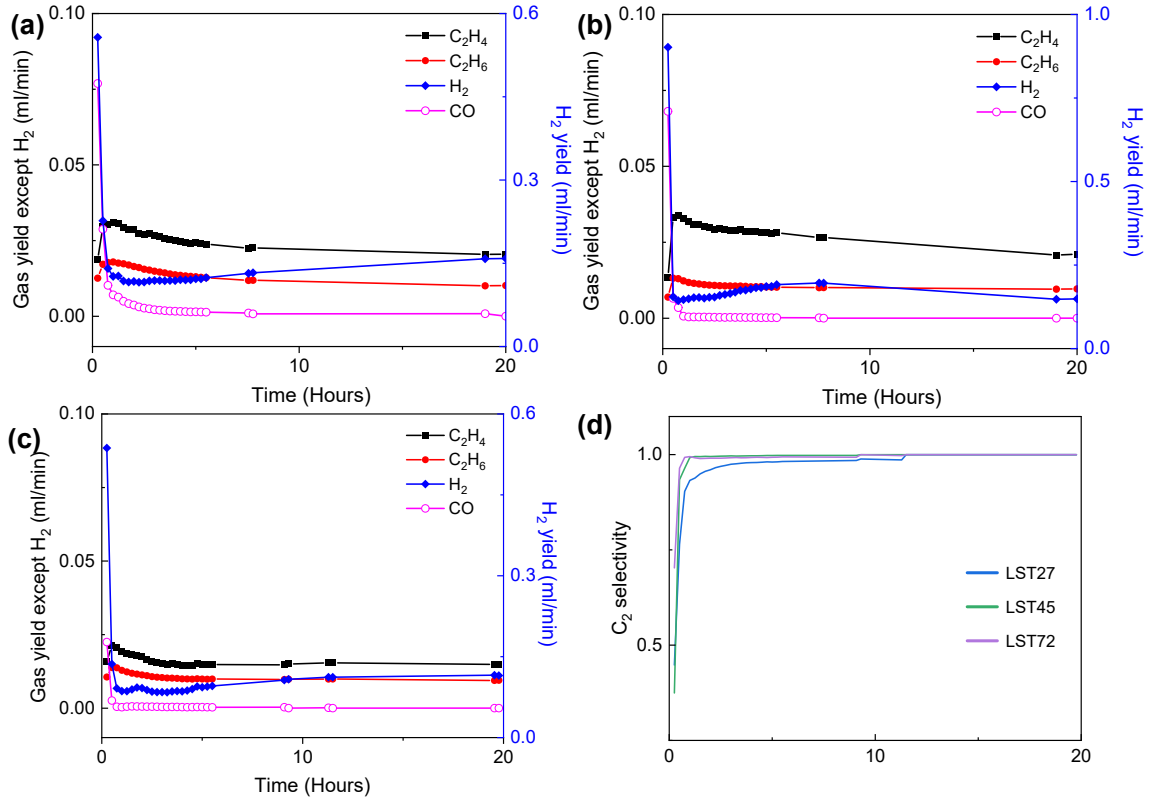

Fig. S3 Total concentration of hydrogen and CO during nonoxidative methane coupling at 850 °C with (a)LST27GDC; (b)LST45GDC; (c)LST72GDC; (d) C_2 selectivity.

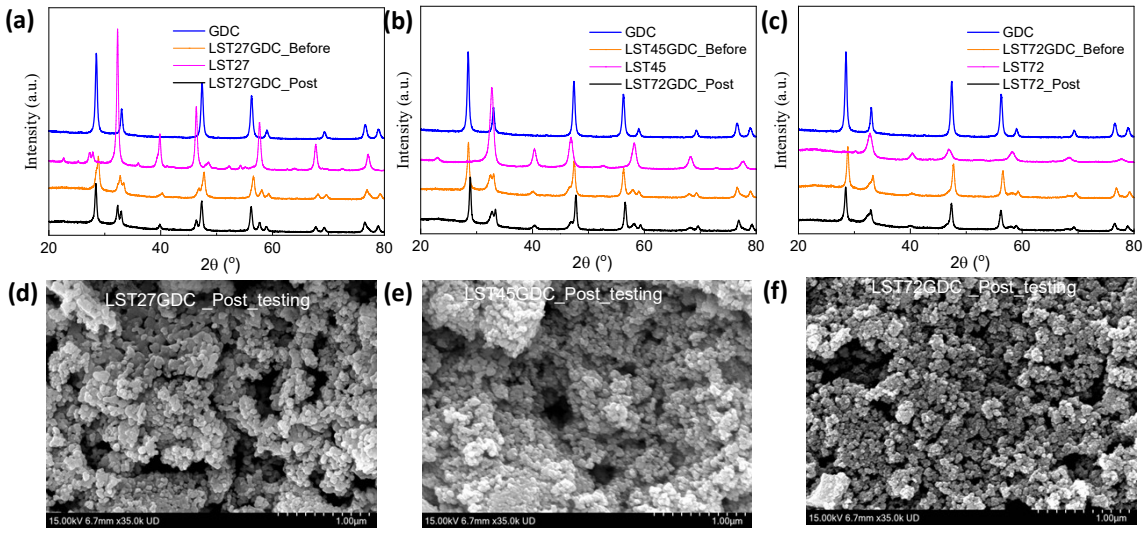

Fig. S4 XRD patters of the samples before testing (a) LST27; (b) LST45 and (c) LST72, SEM images post testing (d) LST27GDC; (e) LST45GDC; (f) LST72GDC.

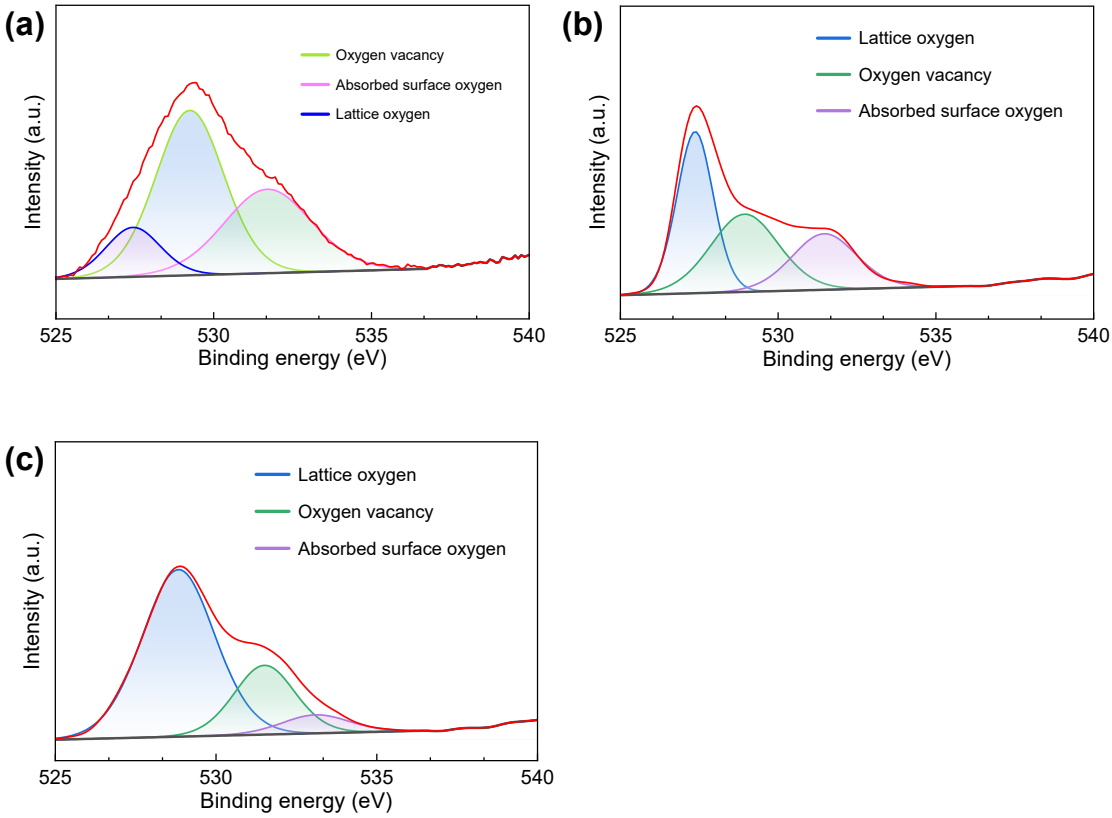

Fig. S5 O1sXPS spectra of (a) LST27 after methane coupling; (b) LST45 after methane coupling; (c) LST72 after methane coupling.

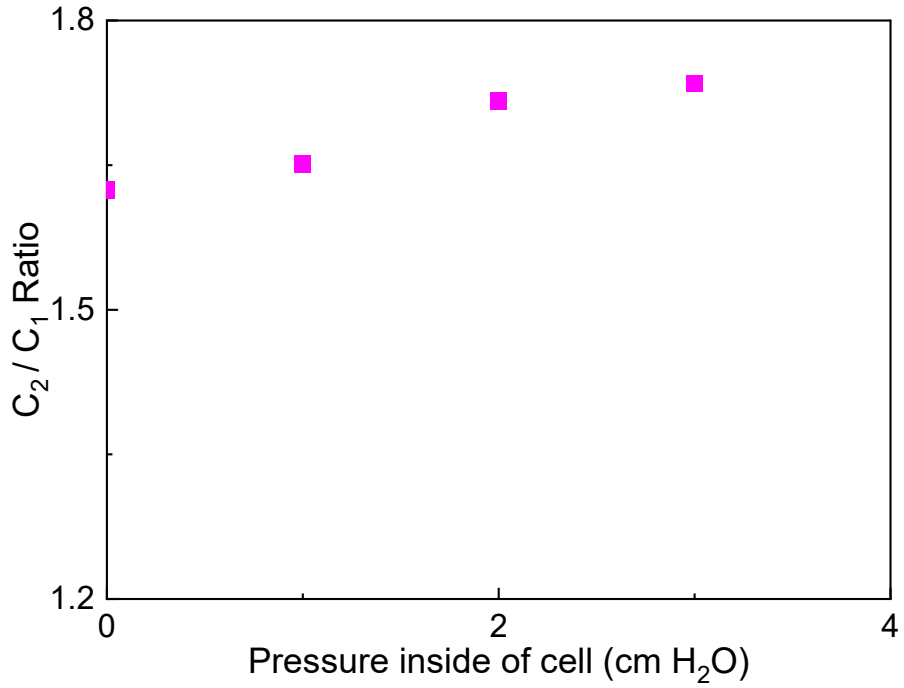

Fig. S6 C_2/C_1 ratio as a function of positive pressure inside of anode chamber (cm H_2O),100ml/m air and 50ml/m CH_4 at 0 current.

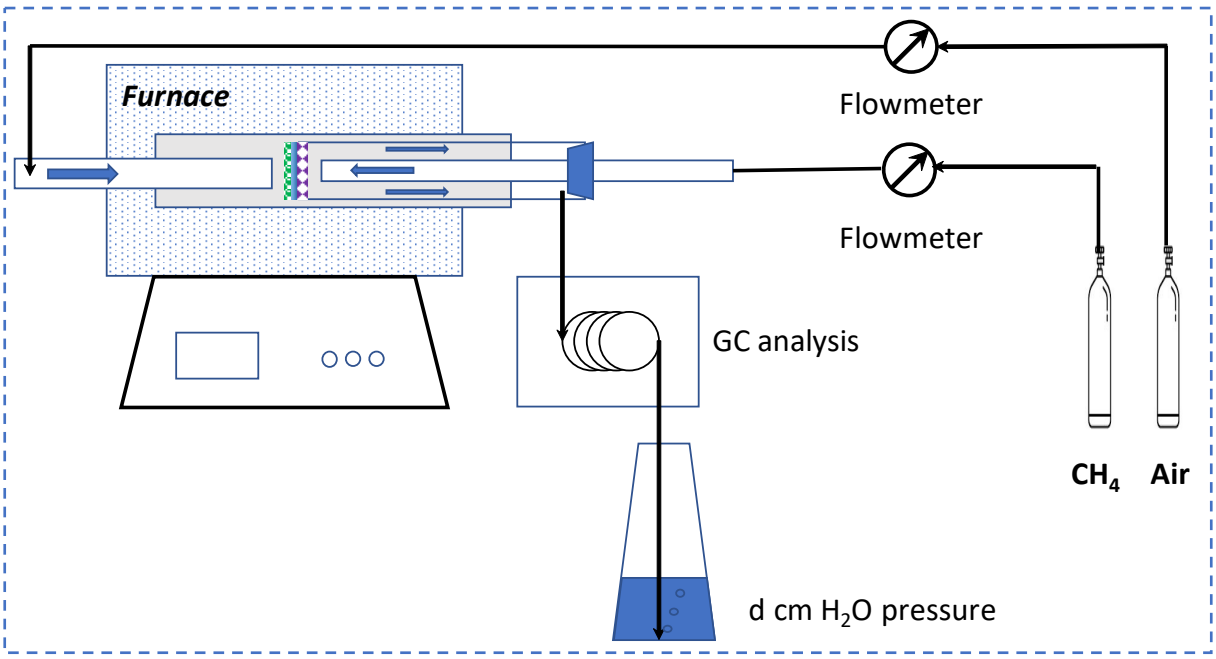

Fig. S7 Schematic of the experimental setup.

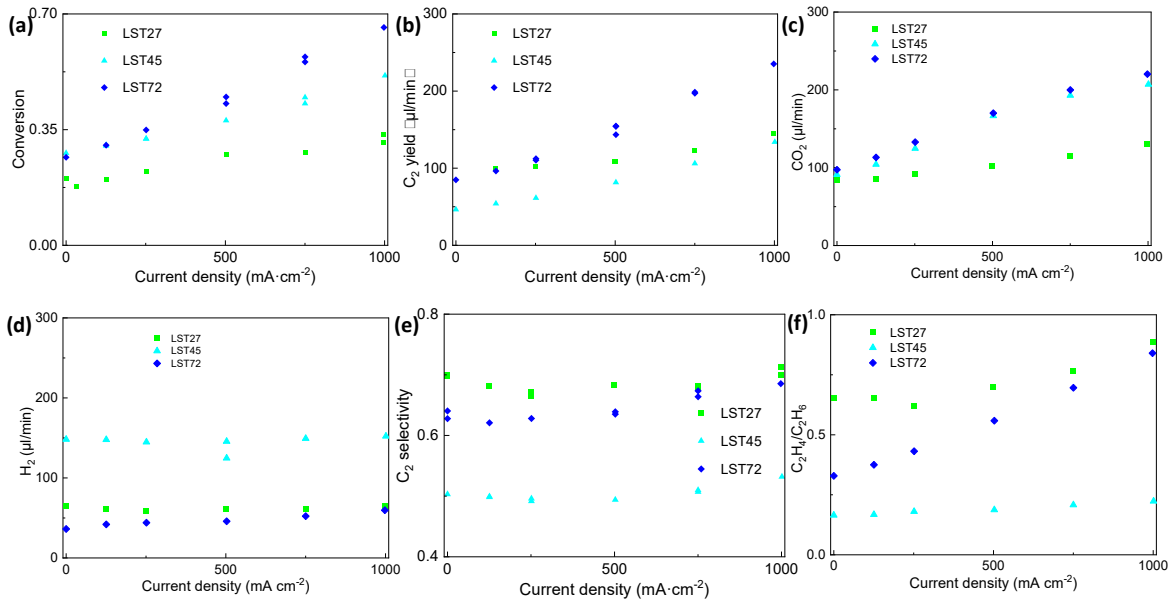

Fig. S8 Catalytic performance for methane conversion using a SOFC button cell design (a) C_2 concentration, (b) CO_2 concentration, (c) H_2 concentration, (d) C_2H_4/C_2H_6 and (e) C2 selectivity and (f) methane conversion% as a function of current density.

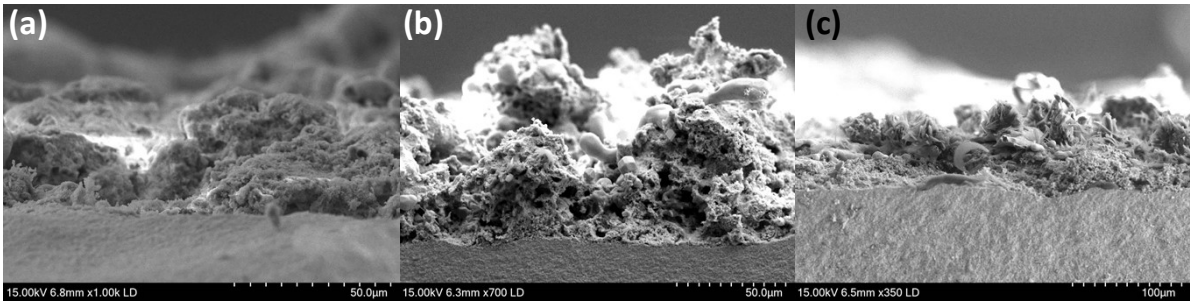

Fig. S9 Cross-sectional images of (a) LST27GDC/GDC, (b) LST45GDC/GDC and (c) LST72GDC/GDC.

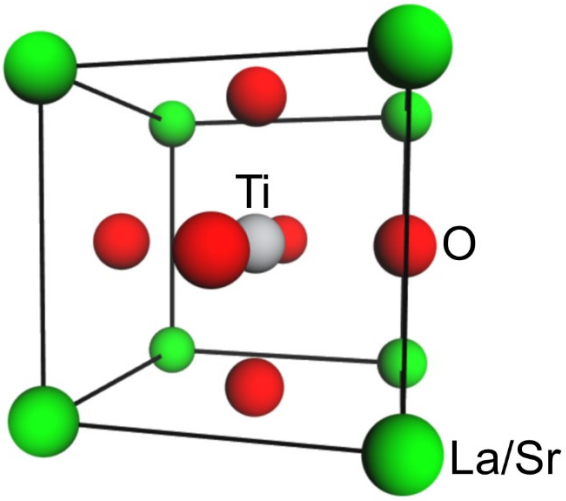

Fig. S10. Crystal structure employed for computational modeling. The structure is based on $SrTiO_3$ crystal, in which the Sr positions are replaced by La in different proportions. La/Sr: large green spheres; Ti: large grey spheres; O: large red spheres.

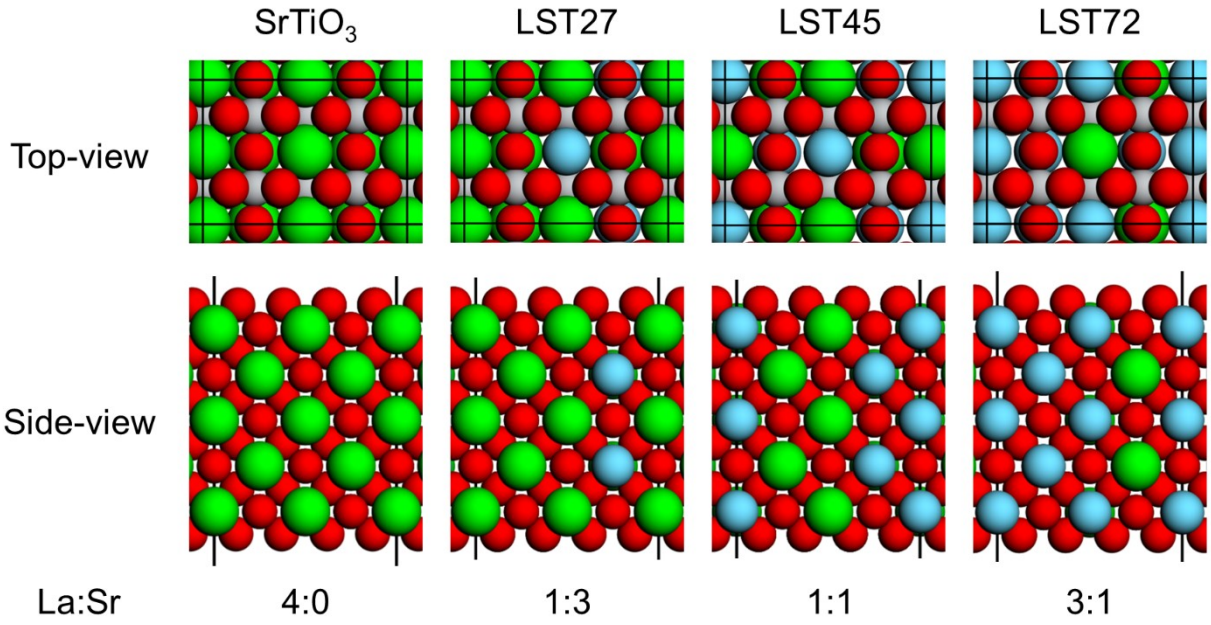

Fig. S11. Computational models of different catalyst surfaces based on $SrTiO_3$(110). For the simulation of LST27, LST45, and LST72 catalysts, a symmetric model is employed where each La/Sr layer follows La:Sr ratios of 1:3, 1:1, and 3:1, respectively. La: large blue spheres; Sr: large green spheres; Ti: large grey spheres; O: large red spheres.

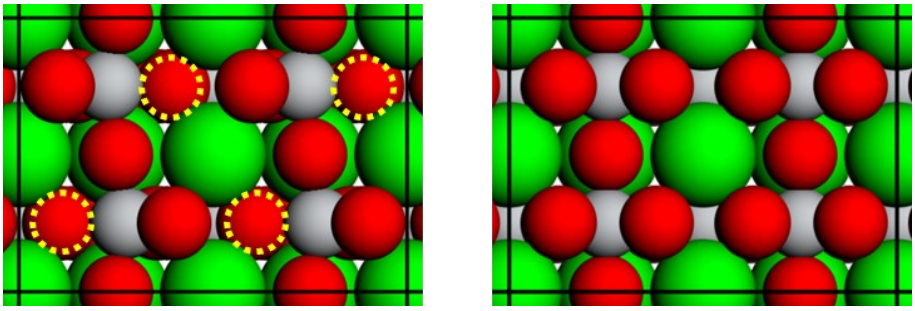

Fig. S12. Surface structures (top view) with (left) and without (right) oxygen (O)-vacancy for calculation of reaction energy (ΔE). The positions of O-vacancies are marked with yellow dotted circles. La/Sr: large green spheres; Ti: large grey spheres; O: large red spheres.