Sulfur tolerance mechanisms of the samarium (Sm)-doped cerium oxide (CeO_2) from first-principles

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The modification of Ni-based anode by CeO_2-based materials has been considered a promising candidate for reducing sulfur poisoning since the oxygen storage capability (OSC) of CeO_2-based materials is expected to greatly enhance the sulfur tolerance. However, the underlying mechanism that governs the sulfur tolerance of CeO_2-based materials still remains unclear. The current study investigates the role of samarium (Sm) in determining the sulfur tolerance of Sm doped-CeO_2 ($Sm_xCe_{1-x}O_2$) using density functional theory calculations. For this, we examined how H_2S is decomposed and SO_2 is formed on $Sm_xCe_{1-x}O_2$ and CeO_2, and analyzed the electronic structures of the surfaces to better understand the origin of sulfur tolerance in CeO_2-based materials. We demonstrate that Sm doping depletes electron density of lattice oxygen facilitating oxygen vacancy formation and promotes H_2S decomposition and SO_2 formation by stabilizing S and lowering SO_2 desorption energy. The enhanced S interaction on Sm doped-CeO_2 is attributed to the Sm 4*f* orbital that fills O 2*p*-Ce 4*f* gap of CeO_2.