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The role of subsurface oxygen in the selectivity enhancement of ethylene epoxidation on Ag-Cu Catalysts NGOC LINH NGUYEN, STEFANO DE GIRONCOLI, International School for Advanced Studies (SISSA), via Bonomea 265 Trieste Italy, SIMONE PICCININ, CNR-IOM, DEMOCRITOS National Simulation Center, Theory@Elettra Group, Trieste, Italy — The role of subsurface oxygen on the Ag-Cu alloy catalysts for the ethylene epoxidation reaction has been studied by means of first principles Density Functional Theory (DFT) calculations. We find that in presence of oxygen and ethylene reactants, the subsurface oxygen adsorption is energetically favorable on fcc sites under the thin oxide-like CuO layer formed at the catalyst surface. On this substrate the reaction proceeds via the formation of a common oxametallacycle precursor. The calculated activation energies show favorable energetics for the pathway leading to the formation of the desired product, ethylene oxide, with respect to the one leading to the formation of the undesired product, acetaldehyde, while the opposite order is obtained on pure Ag catalyst. These findings provide an understanding, at the atomistic level, of the selectivity enhancement of Ag-Cu alloy with respect to pure Ag catalysts. Furthermore, we find that under temperature and partial pressure conditions close to the experimental ones, the ethylenedioxy intermediate is present on the phase diagram of Ag-Cu (111) surface. Our calculations indicate, however, that the formation of this structure could poison the catalyst surface.

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