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First-principles investigation of Ag-Cu alloy surfaces in an oxidizing environment SIMONE PICCININ, CATHERINE STAMPFL, The School of Physics, The University of Sydney, Australia, MATTHIAS SCHEFFLER, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Germany — By means of density-functional theory, together with concepts from atomistic thermodynamics, we present a theoretical procedure for describing the structure and stoichiometry of a binary alloy in contact with a surrounding gas phase environment. We apply the approach to the Ag-Cu alloy in an oxygen atmosphere, for which recent results report a superior selectivity for ethylene epoxidation compared to pure silver, the predominant catalyst for this reaction. We first show that the presence of oxygen leads to copper segregation to the surface. Then, considering the surface free energy as a function of the surface Cu composition, we construct the "convex hull". By including the dependence of the surface free energy of the oxygen chemical potential, we determine the phase diagram of the alloy as a function of temperature, pressure, and Cu surface content. We predict that for conditions typical of the epoxidation reaction, a number of structures can be present on the surface of the alloy including the clean silver surface, thin copper-oxide-like structures, and thick copper oxides. These findings are consistent with, and help explain the recent experimental results. We envisage this approach will be useful and generally applicable for the study of other alloys in contact with a gas or liquid phase.

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